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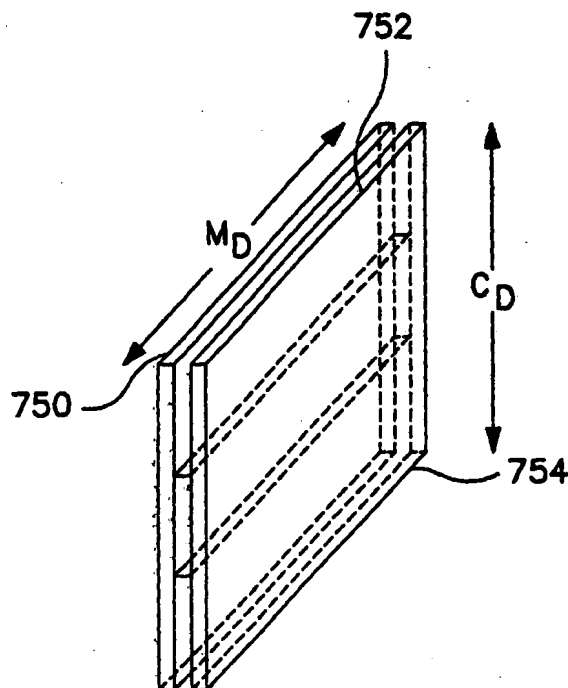
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(54) Title: **HOT-MELT ADHESIVE BASED ON BLEND OF AMORPHOUS AND CRYSTALLINE POLYMERS FOR MULTI-LAYER BONDING**



(57) Abstract: The present invention is generally directed to adhesive compositions comprising selected ratios of crystalline and amorphous polymers. In some versions of the invention, polymers capable of existing in different configurations (*e.g.*, a polymer such as polypropylene which can exist in an atactic, syndiotactic, or isotactic configuration) is used to prepare adhesives of the present invention. As an example, a selected amount of isotactic polypropylene is blended with a selected amount of atactic polypropylene to prepare an adhesive composition having one or more performance properties (*e.g.*, bond strength) that are superior to the performance properties of a conventional hot-melt adhesive.

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HOT-MELT ADHESIVE BASED ON BLEND OF AMORPHOUS AND CRYSTALLINE POLYMERS FOR MULTILAYER BONDING

BACKGROUND OF THE INVENTION

5 People rely on disposable absorbent articles to make their lives easier. Disposable absorbent articles, such as adult incontinence articles and diapers, are generally manufactured by combining several components. These components typically include a liquid-permeable topsheet; a liquid-impermeable backsheet attached to the topsheet; and an absorbent core located between the topsheet and the backsheet. When the disposable article is worn, the liquid-permeable topsheet is positioned next to the body of the wearer. The topsheet allows passage of bodily fluids into the absorbent core. The liquid-impermeable backsheet helps prevent leakage of fluids held in the absorbent core. The absorbent core generally is designed to have desirable physical properties, e.g. a high absorbent capacity and high absorption rate, so that bodily fluids can be transported from the skin of the wearer into the disposable absorbent article.

15 Frequently one or more components of a disposable absorbent article are adhesively bonded together. For example, adhesives have been used to bond individual layers of the absorbent article, such as the topsheet (also known as, for example, the body-side liner) and backsheet (also known as, for example, the outer cover), together. Adhesive has also been used to bond discrete pieces, such as fasteners and leg elastics, to the article. In many cases, the bonding together of components forms a laminated structure in which adhesive is sandwiched between materials (such as layers of polymer film and/or layers of woven or nonwoven fabrics) that make up the components being bonded together.

20 In many instances, a hot-melt adhesive, *i.e.* a polymeric formulation that is heated to substantially liquefy the formulation prior to application to one or both materials when making a laminate, is used in making a laminated structure. While such formulations generally work, they can be costly and their performance properties can be improved. For example, adhesion can be improved to help provide a sturdier laminate (*e.g.*, to improve the integrity or strength of the bond between two components in a disposable absorbent article). Alternatively, the propensity of hot-melt adhesives to migrate from the laminated structure

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and onto the surfaces of equipment, such as ultrasonic-bonding equipment used to ultrasonically bond layers, can be reduced or eliminated.

There is a need or desire for an adhesive composition that possesses one or more performance characteristics that are comparable to, or better than, one or more of the same performance characteristics (*e.g.*, bond strength) of a conventional hot-melt adhesive and that will typically cost less than a conventional hot-melt adhesive. Laminated structures and disposable absorbent articles employing the adhesive composition would benefit from these improved characteristics. There is also a need or desire for efficient methods of making the adhesive composition, and efficient methods of making laminated structures and disposable absorbent articles employing the adhesive composition.

SUMMARY OF THE INVENTION

The present invention is generally directed to adhesive compositions comprising selected ratios of crystalline and amorphous polymers, the adhesive compositions having better performance characteristics and/or costing less than conventional hot-melt adhesives. For example, we have discovered that the performance characteristics of an adhesive composition comprising a polymer which can assume different configurations (*e.g.*, an atactic, isotactic, and/or syndiotactic configuration, as defined below) can be improved by manipulating the ratio of the configurations present in the adhesive composition (*e.g.*, by increasing the amount of a polymer having an isotactic configuration, which typically has a higher degree of crystallinity compared to the other configurations, relative to the amount of polymer having an atactic configuration, which typically has a lower degree of crystallinity compared to the other configurations). So, for example, we have found that a material comprising a combination of atactic polypropylene and isotactic polypropylene possesses desirable adhesive properties and may be used to make laminated structures and disposable absorbent articles.

An atactic, or amorphous, polymer is generally less likely to assume a crystalline structure. An isotactic polymer is generally more likely to assume a crystalline structure. Without being bound to any particular theory, it is believed that a material comprising a specified combination of atactic and isotactic polymers, such as atactic and isotactic polypropylene, possesses regions, and/or characteristics, of both a crystalline

material and an amorphous material. By changing the relative amounts of atactic and isotactic polymer, or for that matter the relative amounts of polymer having differing degrees of crystallinity, one can change the performance characteristics of the resulting adhesive composition. The adhesive compositions of the present invention generally perform better, and cost less, than conventional hot-melt adhesives. It should be understood, however, that the present invention encompasses adhesive compositions comprising selected polymers having different degrees of crystallinity, such as an adhesive composition comprising atactic and isotactic polypropylene, whether or not the composition possesses all of the advantages discussed herein.

As stated above, a material comprising a combination of an atactic and isotactic polymer, such as a material comprising atactic polypropylene and isotactic polypropylene, may cost less than a conventional hot-melt adhesive. Generally this is because conventional hot-melt adhesives are typically formulated by combining several components, including a polymer or polymers for cohesive strength; resins, tackifiers, or other generally low molecular-weight materials for adhesive strength; viscosity modifiers such as oils or wax-like materials; and other additives (*e.g.*, antioxidants). In some versions of the invention, a combination of the atactic and isotactic polymer alone provides improved bond characteristics compared to conventional hot-melt adhesives. But it should be understood that the present invention encompasses adhesive compositions that include selected atactic, isotactic, and/or syndiotactic polymers, or selected polymers having different degrees of crystallinity, combined with other additives or materials.

Another advantage present in some versions of the present invention is that the compositions may be used in conventional hot-melt-adhesive processing equipment. Thus the adhesive material may be used in equipment already installed for the purpose of processing and applying conventional hot-melt adhesives.

Apart from whether or not adhesive compositions of the present invention cost less than conventional hot-melt adhesives, we have found that representative embodiments of our present invention possess improved performance characteristics compared to the performance characteristics of conventional hot-melt adhesives. These performance benefits may justify processing and applying adhesive compositions of the present invention in

modified conventional-hot-melt-adhesive equipment, or in equipment especially designed and built for the purpose of processing and applying adhesive compositions of the present invention. Furthermore, these performance benefits may justify adhesive compositions of the present invention, in some instances, being at a higher cost than conventional-hot-melt adhesives.

A look at one set of results presented in the Examples below demonstrates one advantage that is present in at least some versions of the present invention. In one example, 8 different adhesives, one of which was an example of the present invention, were used to bond two substrates together. All of the test laminates were made using conventional hot-melt processing equipment in which both the conventional hot-melt adhesives and an adhesive of the present invention were substantially liquefied at a temperature ranging from about 350 degrees Fahrenheit to about 380 degrees Fahrenheit. The substantially liquefied adhesive was then conducted from the hot-melt tank, by a gear pump, to a point of application proximate to one of the substrates used to make the laminate. Each of the 8 laminates employing a different adhesive composition were then subjected to a test designed to evaluate the integrity or strength of the bond between the two substrates (again, described in more detail below). The 7 laminates each comprising a different, conventional hot-melt adhesive failed in about 4 hours or less (failure meaning that the two substrates, initially joined using the adhesive, became completely detached from one another; this time of failure is identified as the static-peel-failure time). The laminate employing an adhesive composition of the present invention did not exhibit a bonding failure after 30 hours. Instead, one of the substrates itself failed during the test; *i.e.*, the bond did not fail, there was a failure of the material because the bond was stronger than the material.

One version of an adhesive composition possessing features of the present invention comprises an atactic polymer having a degree of crystallinity of about 20% or less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically about 3000 to about 100,000; and an isotactic polymer having a degree of crystallinity of about 40% or more, specifically of about 60% or more, particularly of about 80% or more, and a number-average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; wherein

the adhesive composition is hot-melt processable at a temperature of about 450 degrees Fahrenheit or less, specifically at a temperature of about 400 degrees Fahrenheit or less, particularly at a temperature of about 375 degrees Fahrenheit or less, and suitably at a temperature of about 350 degrees Fahrenheit or less.

5 This adhesive composition can have a melt index between about 100 and about 2000 grams per 10 minutes, or between about 200 and about 1800 grams per 10 minutes, or between about 500 and about 1500 grams per 10 minutes, as determined using ASTM D 1238. The melt index is dependent upon the crystallinity, molecular weight, and the molecular weight distribution of the polymers included in the adhesive composition. In
10 some versions of the invention, the atactic polymer is present in an amount of about 50 to about 90 weight percent and the isotactic polymer is present in an amount of about 5 to about 50 weight percent. The concept of an adhesive composition being hot-melt processable is discussed in more detail below. The atactic polymer may be the same as the isotactic polymer (*e.g.*, both may be polypropylene, as described below, or both may be polystyrene,
15 polybutene, polyethylene, or combinations of any of these, for example), or the atactic polymer may be different from the isotactic polymer. The term "high density polyethylene" (HDPE) is used to refer to polyethylene that is essentially isotactic, while the term "low density polyethylene" (LDPE) is used to refer to polyethylene that is essentially atactic. HDPE generally has a density in a range of about 0.935 to 0.980 grams per cubic centimeter,
20 while LDPE generally has a density in a range of about 0.910 to 0.935 grams per cubic centimeter.

For purposes of this application, weight percent is defined as the mass of one type of polymer (*e.g.*, atactic) in the adhesive composition divided by the sum of the masses of other types of polymer (*e.g.*, atactic and isotactic) in the adhesive composition, plus the
25 mass(es) of any additional component(s) that might be present in the adhesive composition, with this value being multiplied by 100. So, for example, if we form an adhesive composition comprising 40 grams of atactic polypropylene with 60 grams of isotactic polypropylene, the combination includes 40 weight percent atactic polypropylene.

One version of an adhesive composition possessing features of the present
30 invention comprises atactic polypropylene having a degree of crystallinity of about 20% or

less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically about 3000 to about 100,000; and isotactic polypropylene having a degree of crystallinity of about 40% or more, specifically of about 60% or more, particularly of about 80% or more, and a weight average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; wherein the adhesive composition is hot-melt processable at a temperature of about 450 degrees Fahrenheit or less. This adhesive composition can have a melt index between about 100 and about 2000 grams per 10 minutes, or between about 200 and about 1800 grams per 10 minutes, or between about 500 and about 1500 grams per 10 minutes. Another version of an adhesive composition of the present invention provides that the adhesive composition is hot-melt processable at a temperature of about 400 degrees Fahrenheit or less, specifically at a temperature of about 375 degrees Fahrenheit or less, and more specifically at a temperature of about 350 degrees Fahrenheit or less. Still another version of an adhesive composition of the present invention provides that the atactic polypropylene is present in amount of about 50 to about 90 weight percent and the isotactic polypropylene is present in an amount of about 5 to about 50 weight percent.

In another aspect, the invention encompasses laminated structures employing versions of the adhesive composition as described above. For example, one version of a laminated structure of the present invention comprises a first layer and a second layer, wherein at least a portion of the first layer is attached to at least a portion of the second layer using an adhesive composition that is the same as, or analogous to, one or more of the versions described above, and wherein the laminated structure has a static-peel-failure time of at least about 1 hour, specifically of at least about 8 hours, and particularly of at least about 24 hours.

In yet another aspect, a laminated structure of the present invention comprises a first layer and a second layer, wherein at least a portion of the first layer is attached to at least a portion of the second layer using an adhesive composition that is the same as, or analogous to, one or more of the versions described above, and wherein the laminated structure has a relative accretion value of less than 1, or less than 0.5, or less than 0.2 (or, alternatively, an accretion value that is substantially zero, or an accretion value that is less

than the accretion value of a conventional hot-melt adhesive for which an adhesive composition of the present invention is substituted). A relative accretion value of less than 1 means that the adhesive composition of the present invention builds up on processing equipment, such as ultrasonic-bonding equipment, at a rate, or in an amount, less than a conventional hot-melt adhesive that is selected as the comparator. In some versions of the invention, a laminated structure employing an adhesive composition having features of the present invention, when passed through a unit operation in which the laminated structure is exposed to energy (*e.g.*, ultrasonic energy, infrared energy, thermal energy by conductive or convective transport, and/or the like), produces substantially no build up of the adhesive composition on surfaces of equipment that make up that unit operation (*e.g.*, the surfaces of ultrasonic-bonding equipment used to ultrasonically bond materials).

For any of the laminated structures described above, the first and second layer may be part of one-and-the-same substrate. That is, the substrate may be folded over and joined to itself using an adhesive composition of the present invention.

Furthermore, the first layer, second layer, or both may comprise a variety of materials, including, but not limited to a nonwoven (*e.g.*, a necked-bonded laminate or a spun-bond material); a film; a woven material; an elasticized component; a substrate comprising cellulosic material, thermoplastic material, or both; some combination of these; or the like.

In yet another aspect, an absorbent article may be formed that employs an adhesive composition of the present invention and/or a laminated structure of the present invention. So, for example, one version of an absorbent article of the present invention comprises a liquid-permeable topsheet; a liquid-impermeable backsheet; and a laminated structure having features of the present invention, such as one or more of the versions described above. Some or all of the backsheet may include the laminated structure; some or all of the topsheet may include the laminated structure; the laminated structure may be attached, directly or indirectly, to the backsheet, the topsheet, or both; or a laminated structure or structures may be present in some combination of these.

In addition to various versions of adhesive compositions, laminated structures, and absorbent products of the present invention, the present invention also encompasses methods of making these compositions, structures, and articles of manufacture.

One version of a method of making a laminated structure having features of the present invention comprises the steps of providing a first substrate; providing a second substrate; providing an atactic polymer having a degree of crystallinity of about 20% or less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically about 3000 to about 100,000; providing an isotactic polymer having a degree of crystallinity of about 40% or more, specifically of about 60% or more, particularly of about 80% or more, and a number-average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; heating the atactic polymer and the isotactic polymer so that they are sufficiently liquefied for blending; blending the heated atactic polymer and the heated isotactic polymer to form an adhesive composition that is melt-processable at a temperature of less than about 450 degrees Fahrenheit, specifically of less than about 400 degrees Fahrenheit, particularly of less than about 375 degrees Fahrenheit, and suitably of less than about 350 degrees Fahrenheit; applying the adhesive composition to the first substrate, the second substrate, or both substrates; and joining at least a portion of the first substrate to at least a portion of the second substrate so that some or all of the applied adhesive composition is positioned between the first substrate and second substrate. In some methods of the present invention, the atactic polymer is present in an amount of about 50 to about 90 weight percent and the isotactic polymer is present in an amount of about 5 to about 50 weight percent. The isotactic polymer and the atactic polymer may be the same polymer (*e.g.*, atactic polypropylene and isotactic polypropylene, or atactic polystyrene and isotactic polystyrene, or atactic polybutene and isotactic polybutene, or HDPE and LDPE, or combinations of the same polymers), or different polymers.

It should be understood that the atactic and isotactic polymers could be heated and blended at a site other than the site where the laminate is being formed. For example, atactic and isotactic polymer could be blended using an extruder or hot-melt processing equipment at a first geographic location. The blend could then be allowed to cool and

processed to make a solid form (*e.g.*, pellets). The atactic/isotactic polymer blend, in solid form, could then be shipped from the first geographic site to a site where a laminate is to be made. The blend, in solid form, would simply be heated to substantially liquefy the adhesive composition prior to its being used to make a laminate.

5 It should also be understood that a method having features of the present invention encompasses different sequences of steps by which the adhesive composition is made. For example, the atactic polymer could be heated in a first container; the isotactic polymer could be heated in a second container, before, after, or concurrently with the heating of the atactic polymer; and then the two substantially liquefied polymers could be blended
10 in the first container, the second container, or a third container. Alternatively, one of an atactic or isotactic polymer could be heated in a container, and after the selected polymer was substantially liquefied, the remaining polymer could be added to the same container to be heated and blended. In another alternative, the atactic and isotactic polymer could be added
15 to the same container to be heated and blended at the same time. In other words, our invention contemplates various methods and sequences by which selected amounts of polymer having an atactic, isotactic, and/or syndiotactic configuration (plus any other optional additives), or selected amounts of polymers having different degrees of crystallinity, are heated and blended to form an adhesive composition of the present invention.

 The preceding discussion assumes that the atactic polymer and isotactic
20 polymer are in substantially solid form at room temperature, or temperatures that are typically present in a working environment suitable for human beings. To the extent that the atactic polymer or isotactic polymer is available in substantially liquid form, then those steps providing for heating and liquefying that material (*i.e.*, the already-liquefied material) can be omitted from methods of the present invention.

25 Another version of a method of making a laminated structure having features of the present invention comprises the steps of providing a first substrate; providing a second substrate; providing atactic polypropylene having a degree of crystallinity of about 20% or less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically about 3000 to about 100,000; providing
30 isotactic polypropylene having a degree of crystallinity of about 40% or more, specifically

of about 60% or more, particularly of about 80% or more, and a number-average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; heating the atactic polypropylene and the isotactic polypropylene so that they are sufficiently liquefied for blending; blending the heated atactic polypropylene and the heated isotactic polypropylene to form an adhesive composition that is melt-processable at a temperature of less than about 450 degrees Fahrenheit, specifically of less than about 400 degrees Fahrenheit, particularly of less than about 375 degrees Fahrenheit, and suitably of less than about 350 degrees Fahrenheit; applying the adhesive composition to the first substrate, the second substrate, or both substrates; and joining at least a portion of the first substrate to at least a portion of the second substrate so that some or all of the applied adhesive composition is positioned between the first substrate and second substrate. In some methods of the present invention, atactic polypropylene is present in amount of about 50 to about 90 weight percent and isotactic polypropylene is present in an amount of about 5 to about 50 weight percent.

Again, as mentioned above, the present invention encompasses different sequences of steps and unit operations for making an adhesive composition of the present invention.

Furthermore, the methods described above encompass the first layer, second layer, or both comprising a variety of materials, including, but not limited to a nonwoven (e.g., a neck-bonded laminate or a spun-bond material), a film, a woven material, an elasticized component, a substrate comprising cellulosic material and/or a thermoplastic material, some combination of these, or the like. Also, methods of making a laminated structure of the present invention encompass the first and second layer being part of one-and-the-same substrate. That is, the substrate may be folded over and joined to itself using an adhesive composition of the present invention.

A method of making an adhesive composition having features of the present invention comprises the steps of: providing atactic polymer having a degree of crystallinity of about 20% or less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically of from about 3000 to about 100,000; providing isotactic polymer having a degree of crystallinity of about 40% or

more, specifically of about 60% or more, particularly of about 80% or more, a number-average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; heating the atactic polymer and the isotactic polymer so that they are sufficiently liquefied for blending; and blending the heated atactic polymer and the heated isotactic polymer to form an adhesive composition that is melt-processable at a temperature of less than about 450 degrees Fahrenheit, specifically of less than about 400 degrees Fahrenheit, particularly of less than about 375 degrees Fahrenheit, and suitably of less than about 350 degrees Fahrenheit. In some methods of the present invention, the atactic polymer is present in amount of about 50 to about 90 weight percent and the isotactic polymer is present in an amount of about 5 to about 50 weight percent. The isotactic polymer and the atactic polymer may be the same polymer (*e.g.*, atactic polypropylene and isotactic polypropylene, or atactic polystyrene and isotactic polystyrene, or atactic polybutene and isotactic polybutene, or HDPE and LDPE), or different polymers. Furthermore, the atactic polymer may also include amorphous polyolefin copolymer, such as butene-ethylene copolymer and/or propylene-ethylene copolymer, for example.

Another method of making an adhesive composition having features of the present invention comprises the steps of: providing atactic polypropylene having a degree of crystallinity of about 20% or less, specifically a crystallinity of about 15% or less, and a number-average molecular weight of from about 1000 to about 300,000, specifically about 3000 to about 100,000; providing isotactic polypropylene having a degree of crystallinity of about 40% or more, specifically of about 60% or more, particularly of about 80% or more, and a number-average molecular weight of from about 3000 to about 200,000, more particularly of about 10,000 to about 100,000; heating the atactic polypropylene and the isotactic polypropylene so that they are sufficiently liquefied for blending; and blending the heated atactic polypropylene and the heated isotactic polypropylene to form an adhesive composition that is melt-processable at a temperature of less than about 450 degrees Fahrenheit, specifically of less than about 400 degrees Fahrenheit, particularly of less than about 375 degrees Fahrenheit, and suitably of less than about 350 degrees Fahrenheit. In some methods of the present invention, the atactic polypropylene is present in amount of

about 50 to about 90 weight percent and the isotactic polypropylene is present in an amount of about 5 to about 50 weight percent.

One version of a method in which an adhesive composition of the present invention is metered or delivered at a desired rate to a unit operation (*e.g.*, a unit operation where the adhesive composition is applied to a substrate or substrates in order to make a laminate) comprises the steps of: determining the amount of adhesive composition being used by the unit operation per unit time; and force-adjusting the volumetric flow rate or the mass flow rate of the adhesive composition so that the amount of adhesive composition being metered or delivered to the unit operation corresponds to the amount of adhesive composition being used by the unit operation per unit time.

These and other features, aspects, and advantages of the present invention will become better understood with regard to the following description, appended claims, and accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 gives symbolic representations of syndiotactic, isotactic, and atactic configurations of a polymer.

Figure 2 gives a visual representation of a fringed-micelle model of a material having both amorphous and crystalline regions.

Figure 3 shows a schematic diagram of one version of a method and apparatus for preparing, processing, and delivering an adhesive composition.

Figure 4A shows one version of a feedback control scheme.

Figure 4B shows one version of a feedforward control scheme.

Figure 5 shows one version of a process control system.

Figure 6 shows one version of a process for making a laminate comprising an adhesive composition.

Figure 7A shows a top view of a portion of one version of a laminate.

Figure 7B shows a sectional, perspective view of a test panel cut from one version of a laminate.

DETAILED DESCRIPTION OF THE PRESENTLY PREFERRED EMBODIMENTS

The present invention is generally directed to adhesive compositions comprising selected ratios of crystalline and amorphous polymers. For example, the present invention encompasses adhesive compositions comprising selected amounts of polymers having different configurations (*e.g.*, a combination of atactic polypropylene and isotactic polypropylene). Adhesive compositions of the present invention generally perform better, and typically cost less, than conventional hot-melt adhesives. Furthermore, these compositions may typically be processed and applied using conventional hot-melt adhesive processing equipment. Generally new equipment will not be necessary to use adhesive compositions of the present invention.

Before describing representative embodiments of the invention, it is useful to define a number of terms for purposes of this application. These definitions are provided to assist the reader of this document.

“Nonwoven” fabric or web means a web having a structure of individual fibers or threads that are interlaid, but not in a regular or identifiable manner as in a knitted fabric. Nonwoven fabrics or webs have been formed from many processes such as, for example, meltblowing processes, spunbonding processes, air laying processes, and bonded carded web processes. The basis weight of nonwoven fabrics is usually expressed in ounces of material per square yard (osy) or grams per square meter (gsm) and the fiber diameters are usually expressed in microns. (Note: to convert from osy to gsm, multiply osy by 33.91.)

“Woven” fabric or web means a fabric or web containing a structure of fibers, filaments, or yarns, which are arranged in an orderly, inter-engaged fashion. Woven fabrics typically contain inter-engaged fibers in a “warp” and “fill” direction. The warp direction corresponds to the length of the fabric while the fill direction corresponds to the width of the fabric. Woven fabrics can be made, for example, on a variety of looms including, but not limited to, shuttle looms, rapier looms, projectile looms, air jet looms, and water jet looms.

“Spunbonded fibers”, or “spunbond fibers”, means small-diameter fibers that are typically formed by extruding molten thermoplastic material as filaments from a plurality of fine capillaries of a spinneret having a circular or other configuration, with the diameter of the extruded filaments then being rapidly reduced as by, for example, in U.S. Patent

4,340,563 to Appel *et al.*, and U.S. Patent 3,692,618 to Dorschner *et al.*, U.S. Patent 3,802,817 to Matsuki *et al.*, U.S. Patents 3,338,992 and 3,341,394 to Kinney, U.S. Patent 3,502,763 to Hartman, U.S. Patent 3,502,538 to Petersen, and U.S. Patent 3,542,615 to Dobo *et al.*, each of which is incorporated by reference in its entirety and in a manner consistent with the present document. Spunbond fibers are quenched and generally not tacky when they are deposited onto a collecting surface. Spunbond fibers are generally continuous and often have average diameters larger than about 7 microns, and more particularly between about 10 and 30 microns. A spunbond material, layer, or substrate comprises spunbonded (or spunbond) fibers.

The term "meltblown fibers" means fibers formed by extruding a molten material, typically thermoplastic in nature, through a plurality of fine, usually circular, die capillaries as molten threads or filaments into converging high-velocity heated gas (*e.g.*, air) streams that attenuate the filaments of molten material to reduce their diameter, which may be to microfiber diameter. Thereafter, the meltblown fibers are carried by the high-velocity gas stream and are deposited on a collecting surface to form a web of randomly dispersed meltblown fibers. Such a process is disclosed for example, in U.S. Patent 3,849,241 to Butin. Meltblown fibers are microfibers which may be continuous or discontinuous, are generally smaller than 10 microns in diameter, and are generally self-bonding when deposited onto a collecting surface.

As used herein, the term "microfibers" means small-diameter fibers having an average diameter not greater than about 100 microns, for example, having a diameter of from about 0.5 microns to about 50 microns, more specifically microfibers may also have an average diameter of from about 1 micron to about 20 microns. Microfibers having an average diameter of about 3 microns or less are commonly referred to as ultra-fine microfibers. A description of an exemplary process of making ultra-fine microfibers may be found in, for example, U.S. Pat. No. 5,213,881, entitled "A Nonwoven Web With Improved Barrier Properties".

"Conventional hot-melt adhesive" means a formulation that generally comprises several components. These components typically include one or more polymers to provide cohesive strength (*e.g.*, aliphatic polyolefins such as poly (ethylene-co-propylene)

copolymer; ethylene vinyl acetate copolymers; styrene-butadiene or styrene-isoprene block copolymers; etc.); a resin or analogous material (sometimes called a tackifier) to provide adhesive strength (*e.g.*, hydrocarbons distilled from petroleum distillates; rosins and/or rosin esters; terpenes derived, for example, from wood or citrus, etc.); perhaps waxes, plasticizers or other materials to modify viscosity (*i.e.*, flowability) (examples of such materials include, but are not limited to, mineral oil, polybutene, paraffin oils, ester oils, and the like); and/or other additives including, but not limited to, antioxidants or other stabilizers. A typical hot-melt adhesive formulation might contain from about 15 to about 35 weight percent cohesive strength polymer or polymers; from about 50 to about 65 weight percent resin or other tackifier or tackifiers; from more than zero to about 30 weight percent plasticizer or other viscosity modifier; and optionally less than about 1 weight percent stabilizer or other additive. It should be understood that other adhesive formulations comprising different weight percentages of these components are possible.

While certain versions of the present invention encompass combinations of atactic and isotactic polymers only (*e.g.*, atactic polypropylene and isotactic polypropylene), it should be understood that other embodiments of the present invention comprise components in addition to combinations of atactic and isotactic polymers (or components in addition to selected amounts of polymers having different degrees of crystallinity).

"Hot-melt processable" means that an adhesive composition may be liquefied using a hot-melt tank (*i.e.*, a system in which the composition is heated so that it is substantially in liquid form; *see, e.g.*, the adhesive-supply unit referred to in the Example 1 below) and transported via a pump (*e.g.*, a gear pump or positive-displacement pump) from the tank to the point of application proximate to a substrate or other material; or to another tank, system, or unit operation (*e.g.*, a separate system, which may include an additional pump or pumps, for delivering the adhesive to the point of application). Hot-melt tanks used to substantially liquefy a hot-melt adhesive typically operate in a range from about 100 degrees Fahrenheit to about 450 degrees Fahrenheit. Generally, at the point of application, the substantially liquefied adhesive composition will pass through a nozzle or bank of nozzles, but may pass through some other mechanical element such as a slot. A hot-melt processable adhesive composition is to be contrasted with a composition that requires a

conventional extruder, and the attendant pressures and temperatures characteristic of an extruder, to liquefy, mix, and/or convey the composition. While a hot-melt tank and pump in a hot-melt processing system can handle adhesive-composition viscosities in a range of up to 50,000 centipoise, an extruder can handle and process adhesive-composition viscosities in a range from about 10,000 centipoise to viscosities of several hundred thousand centipoise. An advantage of some adhesive compositions of the present invention is that said compositions are hot-melt processable; *i.e.*, the combination comprising isotactic and atactic polymers may be substantially liquefied in a hot-melt tank and conveyed to the point of application via a pump. As was stated above, however, some adhesive compositions of the present invention may not possess this particular advantage.

Unless otherwise noted, "laminated structure" or "laminate" means a structure in which one layer, material, component, web, or substrate is adhesively bonded, at least in part, to another layer, material, component, web, or substrate. As stated elsewhere in this application, a layer, material, component, web, or substrate may be folded over and adhesively bonded to itself to form a "laminated structure" or "laminate."

"Polymer", as used herein, generally includes, but is not limited to, homopolymers, copolymers, such as, for example, block, graft, random and alternating copolymers, terpolymers, and blends and modifications thereof. As is explained in this document, polymers may assume different configurations, including isotactic, atactic, and syndiotactic configurations. "Configuration" describes those arrangements of atoms that cannot be altered except by breaking and reforming primary chemical bonds (*i.e.*, covalent bonds). In contrast, "conformation" describes arrangements that can be altered by rotating groups of atoms around single bonds. It should be noted that a single polymer chain may be synthesized such that some portions of the chain have an isotactic configuration and some portions of the chain have an atactic configuration.

A graphic example provides additional detail on the types of configurations mentioned above. If a polymer chain is depicted in a fully-extended, planar, zigzag conformation 1100, the configuration resulting when all the substituent groups R 1102 on the polymer lie above (depicted in Figure 1B) or below (not depicted) the plane of the main chain is called "isotactic". If substituent groups lie alternately above and below the plane the

configuration is called "syndiotactic" (depicted in Figure 1A). And a random sequence of substituents lying above and below the plane is described as an "atactic" configuration (depicted in Figure 1C). As discussed above, a polymer, or a region of a polymer, having an isotactic configuration is more likely to assume characteristics of a crystalline structure.

5 For purposes of this invention, the term "isotactic polymer" refers to a polymer that is at least 60% isotactic, suitably at least 70% isotactic, alternatively at least 80% isotactic. A polymer, or a region of a polymer, having an atactic configuration is more likely to assume characteristics of an amorphous structure. An atactic polymer may assume some crystallinity, but the degree of crystallinity is typically less than 20%, or less than 15%. For

10 purposes of this invention, the term "atactic polymer" refers to a polymer that may not be 100% atactic, but is at least 80% atactic. And a polymer, or a region of a polymer, having a syndiotactic configuration can assume characteristics of a crystalline structure, which is similar to the degree of crystallinity in an isotactic configuration.

In this application, "fringed-micelle model" means a theoretical construct characterizing polymeric structures that have both crystalline 150 and amorphous 152 regions (one version of a graphic depiction of a fringed-micellar structure is presented in Figure 2). This model may be used to characterize the structure of an atactic polymer and an isotactic polymer individually, i.e., each polymer possesses both crystalline regions and amorphous regions. As explained above, the isotactic polymer likely possesses a greater

20 degree of crystallinity compared to an atactic polymer. Furthermore, this model may be used to characterize the structure of a blend of isotactic polymer and atactic polymer. It should be understood that this model provides one possible view of characteristics of the present invention and in no way limits the scope thereof.

The term "open time," as used herein, refers to the length of time during which

25 an adhesive composition remains tacky or sticky prior to drying. Open time is affected by isotacticity of a polymer, such that the greater the level of isotacticity the shorter the open time.

In the process description that follows, the preparation, processing, and application of an adhesive composition comprising atactic polypropylene and isotactic polypropylene is described. It should be understood, however, that this description is given

30

as an example. Other processing methods and equipment may be used to prepare and deliver various adhesive compositions of the present invention.

Figure 3 shows a schematic diagram of an apparatus 20, and a method for spraying an adhesive composition, on a moving web 22. The apparatus 20 may include a programmable control system 24 that is operatively connected to a flow-control system 26. The combination of the programmable control system 24 and the flow-control system 26 are configured to control the delivery of an adhesive composition in liquid form to the moving web 22. Generally an adhesive composition is received in solid form at a manufacturing site where equipment such as that depicted in Figure 3 is located. For example, hot-melt adhesive compositions may be received as solid pellets, blocks, or some other shape. The solid is then heated so that the hot-melt adhesive composition is in a form such that it can be conveyed, and applied, to a substrate or other material. Usually this requires that the heated hot-melt adhesive be in substantially liquid form. For the present invention, an adhesive composition comprising an atactic and isotactic polymer (*e.g.*, atactic polypropylene and isotactic polypropylene), in solid form, might be received at a manufacturing site for heating and processing as described above. Alternatively, the atactic and isotactic polymer might be received as separate components to be blended at the manufacturing site (as discussed above in the Summary, the present invention encompasses a variety of sequences of steps for making adhesive compositions of the present invention). An example of equipment and methods for heating an adhesive composition, or precursor materials to the adhesive composition, are described in more detail below.

The apparatus may also include a position-sensing system that is configured to sense a position of the moving web 22 and, in response thereto, generate a signal that is sent to the programmable control system 24.

As representatively illustrated in Figure 3, the continuously moving web 22 may be supplied by any means known to those skilled in the art, such as known conveyor systems. The continuously moving web 22 can include any type of layer or web of material, such as: films; nonwoven webs; woven webs which may include strands of thermoplastic material; an elasticized component; natural material such as threads of cotton and the like; laminate materials; or combinations thereof. More particularly, the continuously moving

web 22 may include a necked-bonded laminate ("NBL"), which generally comprises a polyethylene layer sandwiched between two polypropylene, spunbonded layers; a polypropylene, spunbonded layer ("SB"); or an outercover comprising a polyethylene layer and a polypropylene, spunbonded layer. As is described below in more specific terms, the adhesive is sprayed on the continuously moving web 22 in a specific design or pattern for subsequent placement of or bonding to another material. The other material can be the same or different than the web to which adhesive was applied. In some cases adhesive might be applied to both substrates before they are joined together. And, as mentioned in the Summary section above, one substrate might be folded over and attached to itself to form a laminated structure.

The programmable control system 24 of the present invention is configured to send signals to the flow control system 26 which, in response thereto, is configured to initiate a spray of adhesive at the correct time to provide the desired pattern of adhesive on the moving web 22. As representatively illustrated in Figure 3, the flow control system 26 includes an adhesive source 28 which is configured to deliver an adhesive through an adhesive supply line 30 to a metering mechanism 32. The adhesive can be delivered to the metering mechanism 32 by any means known to those skilled in the art, such as by the use of a pump.

The metering mechanism 32 is configured to continuously supply at least one independent, volumetric flow of adhesive to a respective nozzle. As used herein, the term "volumetric flow" refers to a flow of adhesive that has a predetermined volumetric flow rate. Such a "volumetric flow" may be provided by a positive-displacement metering pump which is configured to supply a specific volumetric flow which is independent of the manner in which the adhesive is supplied to the metering mechanism 32. As a result, for an adhesive that is at a given density, the metering mechanism 32 is configured to provide an independent, predetermined mass flow rate of adhesive to each nozzle. Other adhesive processing and delivery systems utilize pressure to provide a flow of adhesive.

The metering mechanism 32 of the present invention may be configured to supply a single, volumetric flow of adhesive to one nozzle or an independent, volumetric flow of adhesive to each of a plurality of nozzles depending upon the number of nozzles

required to provide the desired pattern of adhesive on the moving web 22. A suitable device to provide the metering mechanism 32 may include a positive-displacement metering pump which is commercially available from May Coating Technologies, Acumeter Division, a business having offices located in Holliston, Mass., under the trade designation No. 19539. The metering mechanism 32 may include any other piston pump or gear pump which are well known to those skilled in the art.

The metering mechanism 32 may be configured to supply any desired volumetric flow rate of adhesive to each nozzle. For example, the metering mechanism 32 may be configured to provide a pre-determined volumetric flow rate of from about 1 to about 1000 cubic centimeters per minute and suitably from about 30 to about 180 cubic centimeters of adhesive per minute to each nozzle. The metering mechanism 32 may be configured to provide either a constant or a variable volumetric flow rate of adhesive to each nozzle. For example, if the metering mechanism 32 is a positive-displacement metering pump, the speed of the pump may be controlled to vary the volumetric flow rate of adhesive to the nozzles.

Each nozzle 38 and 40 as representatively illustrated in Figure 3 can be any device which is capable of providing the desired pattern of adhesive on the moving web 22. For example, one suitable nozzle is commercially available from Nordson Corporation, a business having offices located in Duluth, Ga., under the trade designation Model No. 144906. Another suitable nozzle for use in the present invention is obtainable from ITW Dynatec Co. of Hendersonville, Tenn., under the trade designation number 057B1639, I.D. #A3. Such nozzles are typically configured to be operated between an on position and an off position to control the spray of adhesive from the nozzles. When operated in the on position, each nozzle may be configured to spray substantially the entire volumetric flow of adhesive which is independently supplied to it to more accurately control the amount and pattern of the adhesive on the moving web. The nozzles 38 and 40 may further be configured to include air streams that can be directed to provide a desired pattern in the spray of adhesive being dispensed from each nozzle. Such air streams can provide a desired adhesive spray pattern, such as a pattern of swirls of adhesive.

After the pattern of adhesive has been sprayed on the moving web 22, the web may be further processed in a variety of ways. For example, the continuously moving web

22 may be contacted by a second substrate web, such as a nonwoven layer, between a pair of nip rolls to adhesively join the two substrate webs together. Thereafter, this composite material or laminate may be used in a variety of ways such as in the construction of disposable absorbent articles such as diapers, incontinent articles, training pants, feminine care articles and the like.

The above discussion provides one example of hot-melt processing equipment and a system for applying adhesive to a substrate. It should be understood that this is but one example, and that the present invention encompasses other systems for preparing and applying adhesives (*see, e.g.*, U.S. Patent No. 4,949,668, entitled "Apparatus for Sprayed Adhesive Diaper Construction," which issued on 21 August 1990, and which is hereby incorporated by reference in its entirety and in a manner consistent with the present document).

Regardless of the system used to apply the adhesive, the resulting composite material or laminate may be exposed to thermal, infrared, ultrasonic, or other forms of energy in subsequent unit operations or processing steps. For example, the laminate or composite material may pass through an ultrasonic-bonding unit operation wherein the laminate or composite material are exposed to ultrasonic energy. After exemplary composite materials or laminates such as those referred to above are formed using an adhesive composition of the present invention, some or all of the composite or laminate may be exposed to ultrasonic energy. Referring to PCT International Publication Number WO 99/25296, which is hereby incorporated by reference in its entirety in a manner consistent with the present document, the publication discloses the use of ultrasonic bonding to form side seams or seals in the disposable underpant. (*See, e.g.*, page 29, lines 10-25; additional detail regarding the forming of such side seals is disclosed in U.S. Patent Number 4,610,681, which issued on 09 September 1986 and is entitled "Disposable Underpants Having Discrete Outer Seals," and which is hereby incorporated by reference in a manner consistent herewith; and U.S. Patent Number 4,641,381, which issued on 10 February 1997 and is entitled "Disposable Underpants, Such as Infant's Training Pants and the Like," which is also incorporated by reference in a manner consistent with the present document.) Thus adhesives of the present invention, used to make laminates and composite materials, may be exposed to ultrasonic

energy when ultrasonic-bonding equipment is used in subsequent processing steps (*e.g.*, when the ultrasonic bonding equipment is used to form the seams or seals in the disposable absorbent article as discussed above).

5 It has been observed that one or more components of some conventional hot-melt adhesives flow when exposed to ultrasonic energy. The flowing adhesive, or components thereof, may then penetrate through the laminate or composite material to contact the surfaces of ultrasonic-bonding equipment. Alternatively, or in addition to, this mechanism, the adhesive, or components thereof, may flow through gaps between a laminate or composite material and build up on the surface of the ultrasonic-bonding equipment. This
10 in turn may lead to build up or accretion of adhesive on parts of the ultrasonic-bonding equipment. As the adhesive builds up, the adhesive may bind other materials such that the build up or accretion comprises both adhesive and other materials, such as fibers from a nonwoven web or other component or piece. This build up or accretion may decrease performance of the ultrasonic-bonding equipment, and may lead to increased down time of
15 the production machine used to make the substrate composite and/or absorbent product.

As is discussed below in the Examples section, conventional hot-melt adhesives or one or more of their components do build up on the surface of ultrasonic-bonding equipment. We have found that some embodiments of adhesive compositions of the present invention either do not build up on the surfaces of ultrasonic-bonding equipment,
20 or, if build-up occurs, the build-up occurs in an amount, or at a rate, that is less than the amount or rate of build-up associated with conventional hot-melt adhesives.

Without being bound to a particular theory, we believe that those embodiments of the present invention that do not contain significant amounts of low molecular-weight materials are less likely to flow when exposed to ultrasonic energy. Significant quantities of
25 smaller molecules (such as a tackifier, resin, oil, or viscosity modifier), when exposed to ultrasonic energy, may assume fluid-like characteristics and migrate. Also, to the extent that the smaller molecules are absorbing ultrasonic energy, the ultrasonic bonding step is less efficient because some of the energy is not being used to weld or join substrates together, but instead is being absorbed by the smaller molecules in the adhesive. Absorption of energy
30 by these smaller molecules results in the adhesive, or part of the adhesive, exhibiting viscous

flow behavior. Accordingly, some embodiments of the present invention, which contain only specified amounts of polymer having an atactic, syndiotactic, and/or isotactic configuration (and optionally containing small amounts of certain additives), do not contain significant quantities of low molecular-weight materials. We believe these embodiments are less susceptible to flowing and building up on the surfaces of equipment when exposed to various forms of energy (e.g., ultrasonic energy when a laminate or composite material comprising the adhesive embodiment is processed in an ultrasonic-bonding unit operation).

In one embodiment of the invention, in addition to including an isotactic polymer and an atactic polymer, the adhesive composition also includes up to 50% by weight of a combination of additives, such as a tackifier, an anti-oxidizing agent, color pigment, filler, and/or a polymer compatibilizer. The adhesive composition may include any one or more of these additives. Examples of suitable tackifiers include PICCOLYTE® S Resins, REGALITE® series, STAYBELITE® ester, each available from Hercules Incorporated, Wilmington, Delaware. The adhesive composition may suitably include 10 to 20 percent by weight tackifier. Examples of suitable anti-oxidizing agents include IRGANOX® 565, available from Ciba-Geigy, POLYGARD®, available from Uniroyal Chemical Co., and ANTIOXIDANT® series, available from Cytec Industries. The adhesive composition may suitably include 0.1 to 1.0 percent by weight antioxidant. Examples of suitable color pigments and fillers include TiO₂, carbon black, and calcium carbonate. The adhesive composition may suitably include 1 to 10 percent by weight color pigments and fillers. Examples of suitable polymer compatibilizers include polypropylene-b-polyethylene, polypropylene-b-polybutene diblock copolymers. The adhesive composition may suitably include 2 to 10 percent by weight polymer compatibilizer. In this embodiment, the adhesive composition suitably has an open time of up to 2 minutes. Alternatively, the adhesive composition can have an open time of up to 30 seconds, or up to 10 seconds, or as short as up to 1 second.

Specific examples of composite materials, laminates, and disposable absorbent articles with which adhesives of the present invention may be utilized are disclosed in the following U.S. Patents and U.S. Patent Applications: U.S. Patent No. 4,798,603 issued Jan. 17, 1989, to Meyer *et al.*; U.S. Patent No. 5,176,668 issued Jan. 5, 1993, to Bernardin;

U.S. Patent No. 5,176,672 issued Jan. 5, 1993, to Bruemmer *et al.*; U.S. Patent No. 5,192,606 issued Mar. 9, 1993, to Proxmire *et al.*; U.S. Patent No. 4,940,464, entitled "Disposable Incontinence Garment or Training Pant"; U.S. Patent No. 5,904,675, entitled "Absorbent Article With Improved Elastic Margins and Containment System"; U.S. Patent No. 5,904,672, entitled "Absorbent Article Having Improved Waist Region Dryness and Method of Manufacture"; and U.S. Patent No. 5,902,297, entitled "Absorbent Article Having a Collection Conduit." Each of the preceding U.S. patents is incorporated by reference in its entirety and in a manner consistent with the present document. It should be understood that the present invention is applicable to other structures, composites, or products incorporating adhesive compositions of the present invention.

Additional Detail on Representative Process-Control Embodiments

As discussed above, process-control systems may be used to control the volumetric or mass flow rate of adhesive compositions of the present invention to a point of application (e.g., to a point of application on a substrate, layer, or web that will be used to make a laminate or composite material). Persons of ordinary skill in the art of process control are familiar with the various process-control strategies, algorithms, and equipment used to control a process. Some of the possible strategies that may be used to control a process include feedback-control strategies (*i.e.*, a process in which a variable to be controlled is measured, the measured value is compared to a desired value, and the difference between the measured value and the desired value is transmitted to a feedback controller that force adjusts a manipulative variable to drive the measured variable back to the desired value) (*see, e.g.*, Figure 4A); feedforward-control strategies (*i.e.*, a process in which a disturbance entering a process is detected, and an appropriate change is made to a manipulative variable so that an output variable is held constant; *see, e.g.*, Figure 4B); and the like.

One example of a process-control system is depicted in Figure 5. A sensor may be used to determine a signal S_1 corresponding to the variable to be controlled, *e.g.* the volumetric or mass flow rate of adhesive being sprayed or delivered in an adhesive-application unit operation 74. This signal may then be relayed electrically, pneumatically,

hydraulically, or by other means to a transmitter 76, which converts the signal S_1 into a control signal M_1 . The transmitter transmits the control signal M_1 to the controller 78.

After receiving the control signal M_1 , the controller sends the corresponding output signal R_1 to the control element 80. The control element, such as an electronic or pneumatic control valve, responds to the output signal R_1 by opening or closing, thus effecting the desired change to the variable being manipulated, in this case the volumetric or mass flow rate of adhesive. Alternatively, the control element might effect a desired change to the speed at which a pump operates, thereby controlling the mass or volumetric flow rate of adhesive.

As mentioned above, an air-pressure, electrical, pneumatic, or other signal may be used to transmit information (*e.g.*, the various signals discussed in the preceding paragraphs) from one device to another (*e.g.*, from a sensor, to a transmitter, to a controller, to a control element, or to some combination of some or all of these). For example, the controller may be a device that converts a control signal into an equivalent air-pressure, electrical, pneumatic, or other output signal. This air-pressure, electrical, pneumatic or other output signal is sent from the controller to a control element that effects a change to the variable being manipulated. If the output signal is an air-pressure signal, the output signal will be transmitted to the control element via tubing. The control element, such as a pneumatic control valve, responds to the output signal by opening or closing, thus effecting the desired change to the variable being manipulated. The control system may include multiple valves: *e.g.*, a two-valve system with one operating as a one-directional, open-or-shut valve and the other operating as a proportional valve. Alternatively, the output signal is converted into an electrical signal. The output signal is relayed to the control element via metal wire or other electrical conductor. The control element, such as an electronic control valve, responds to the electrical signal by opening or closing, thus effecting the desired change to the variable being manipulated.

An operator may input a value directly to the controller to produce a control signal. For example, an operator may adjust a dial or other input device on a pneumatic, hydraulic, electronic, or other controller to adjust the volumetric or mass flow rate of adhesive. The operator selects a setting on the input device of the controller corresponding

to the flow rate desired by the operator. Typically the operator will have calibrated the input device on the controller so that input-device settings each correspond to specific volumetric or mass flow rate values.

5 A general-purpose computer may be used in place of, or in addition to, the controller mentioned above. Typically a general-purpose computer employs an input device, including, but not limited to, an alpha-numeric keyboard, mouse, joystick, stylus, touch screen, or some combination of these. Other devices which may be used to input data to the computer include, but are not limited to: devices for reading data stored on magnetic media such as 3.5 inch "floppy disks" or fixed-drives; devices for reading data stored on optical
10 media, such as CD-ROMs; devices for reading data transmitted over cables, including optical cables; and devices for scanning and digitizing information on a document. In addition to the input devices like those mentioned above, a general-purpose computer usually includes a visual display for displaying data. Also, a general-purpose computer typically has a device for storing and retrieving data that is inputted to the computer. Devices for storing and
15 retrieving data include, but are not limited to: a disk drive for reading data from, and storing data on, a 3.5 inch "floppy disk"; a hard disk or other fixed drive; a tape drive; or other device capable of reading data from, and storing data on, magnetic media.

A general-purpose computer may be adapted for use in controlling the volumetric or mass flow rate of adhesive. Typically a general-purpose computer comprises
20 devices for data input, data storage, data processing, data display, and data output, as discussed above. For purposes of controlling volumetric or mass flow rate, the general-purpose computer may further comprise a set of instructions comprising the following steps: reading the control signal M_1 , the control signal M_1 being transmitted to the computer in computer-readable form; correlating the control signal M_1 to an output signal R_1 and
25 transmitting the output signal R_1 to a control element. The control element, such as an electronic, hydraulic, pneumatic, or other control valve, responds to the output signal R_1 by opening or closing, thus effecting the desired change to the variable being manipulated, in this volumetric or mass flow rate. Alternatively, the control element may effect desired changes to the speed at which a positive-displacement or other metering pump operates,
30 thereby effecting desired changes to mass or volumetric flow rates.

The above discussion provides exemplars of equipment and methods for controlling the amount of adhesive being conducted to a point of application per unit time. It should be understood that other equipment and methods used to force adjust the flow rate of an adhesive of the present invention to a control set point, operator-inputted value, or other desired value falls within the scope of the present invention.

TESTS / PROCEDURES

Laminate Production

Laminates were made on equipment available from J & M Laboratories, a business having offices located in Dawsonville, Georgia. As depicted in Figure 6, a first substrate or first base material 102, such as a nonwoven web, was directed from its corresponding unwind stand (not shown) to the surface of a 6-inch-diameter steel roll 104 and through a nip 106 between the steel roll and a 4-inch-diameter rubber roll 110. A second substrate or second base material, such as a second nonwoven web 108, was directed from its unwind stand (not shown) to the surface of the rubber roll and through the nip. Typically, the equipment was operated at a speed of 300 feet per minute.

The applicator 114 used to deposit the adhesive was positioned so that the face of the depicted nozzle, which was roughly parallel to the surface of the web to which adhesive was first applied, was 1.5 inches 116 from the surface of the web. Furthermore, the central axis of the depicted nozzle, which is perpendicular to the web to which adhesive is first applied, was 8 inches 118 from a parallel axis that passes through the nip defined by the rubber and steel rolls.

From the discussion above, it should be understood that the substrates and the resulting laminate 700 generally moved in a machine direction 702 (*see* Figure 7A) during their preparation. Figure 7A depicts a top view of a portion of a laminate after it has been formed. A continuous band of adhesive 703, whether it was applied using meltblowing, cycloidal, slot, or other application technique, is denoted by broken lines 705 and 707. The adhesive is under the upper substrate of the laminate depicted in the Figure. As the laminate is made in a continuous manner, it is wound up in the form of a roll. The direction that is perpendicular to the machine direction, but lying within the plane of the laminate, is denoted as the cross-machine direction 704. Typically the width of the formed laminate, width

denoting the dimension parallel to the cross-machine direction, was about 1 to about 12 inches 706. The width of the applied adhesive, again width denoting a dimension parallel to the cross-machine direction, typically was from about 0.5 inch to about 10 inches 708. Also, the band of adhesive was generally applied such that it was substantially centered in the laminate (in the width dimension). Unless otherwise noted, the width of the applied adhesive was about 0.5 inch to about 2 inches. (Note: the lines 710 and 712 denote the manner in which a 2-inch 714 sample was cut for subsequent analysis; sample preparation and orientation is discussed in more detail below).

The selected adhesive was either an adhesive of the present invention (as noted in the Examples below), or a hot-melt adhesive (again as noted in the Examples below). The adhesive was added using a variety of patterns, including a meltblown pattern, a swirl or cycloidal pattern, or a pattern resulting from slot coating. Typically the adhesives were heated to temperatures ranging from about 350 degrees Fahrenheit to about 380 degrees Fahrenheit prior to application to one of the substrates. Unless otherwise noted, the selected adhesive was added using a meltblown pattern. As stated above, unless otherwise noted the width of the added adhesive was about 0.5 inch to about 2 inches. The selected adhesive was added in amounts varying from about 1 gram per square meter to about 50 grams per square meter, with specific application levels or add-on levels noted in the examples.

A number of different substrates were used to prepare the laminates, as noted in the Examples below. The substrates that were used included: a necked-bonded laminate ("NBL"), which generally comprised a polyethylene layer sandwiched between two polypropylene, spunbonded layers; a polypropylene, spunbonded layer ("SB"); and an outercover comprising a polyethylene layer and a polypropylene, spunbonded layer. For tests where the performance of a laminate of the present invention was compared to the performance of a laminate prepared using a conventional hot-melt adhesive, the same substrates were used to prepare both the laminate of the present invention and the conventional laminate.

180° Static Peel Test

The static peel test was used to determine the approximate time to failure of a laminate in which one substrate was adhesively bonded to another substrate. All laminates

were made as described above on a J & M machine. Samples were cut from the prepared laminate which was in the form of a continuous web prepared on a J & M machine, as shown in Figure 7A. Figure 7B depicts a sectional view of a sample that has been removed from the laminate depicted in Figure 7A. The test procedure was conducted as follows: 1. A 2-inch test panel was cut from the laminate, as shown in Figures 7A and 7B. 2. The test laminate was then suspended vertically in a forced-air oven, model number OV-490A-2 manufactured by Blue M Co., a business having offices in Blue Island, Illinois, that had been pre-heated to a temperature of 100 degrees Fahrenheit, with the top of one substrate layer secured by a clamp or other mechanical securing element, the clamp or securing element having a width of about 2 inches. 3. A 500-gram weight was then affixed to the top edge of the other substrate using a clamp or other mechanical securing element. Again, the clamp or securing element used to attach the 500-gram weight was about 2 inches. 4. Approximately every ½ hour, the test laminate was visually examined by quickly opening the oven door. The time at which one substrate or layer had detached from the other substrate or layer was recorded. The recorded time corresponded to the approximate time of failure of the laminate. The two, now separate, substrates were then examined to determine the nature of the failure. If the substrates separated such that most of the adhesive remained on one of the substrates, then failure was deemed to be an adhesion failure (*i.e.*, failure likely occurred at the interface between one of the substrates and the adhesive composition). If the substrates separated such that adhesive remained on both substrates, the failure was deemed to be a cohesion failure (*i.e.*, separation likely occurred within the adhesive composition itself). If neither of these conditions arose, but instead one or both of the substrates failed (*i.e.*, that portion of the laminate bonded by the adhesive, usually a 1 inch by 2 inch area of the test panel), then the failure was deemed a material failure of one or both substrates.

Static Shear Test

The static shear test was used to determine the approximate time to failure of a laminate in which one substrate was adhesively bonded to another substrate. All laminates were made as described above on a J & M machine. Samples were cut from the prepared laminate, which was in the form of a continuous web prepared on a J & M machine, as

shown in Figure 7A. Figure 7B depicts a sectional view of a sample that has been removed from the laminate depicted in Figure 7A. The test procedure was conducted as follows: 1. A 2-inch test panel was cut from the laminate, as shown in Figures 7A and 7B. 2. The test laminate was then suspended vertically in a forced-air oven, model number OV-490A-2 manufactured by Blue M Co., a business having offices in Blue Island, Illinois, that had been pre-heated to a temperature of 100 degrees Fahrenheit, with the top of one substrate layer 750 secured by a clamp or other mechanical securing element, the clamp or securing element having a width greater than 2 inches. 3. A 500-gram weight was then affixed to the bottom edge 754 of the other substrate using a clamp or other mechanical securing element. Again, the clamp or securing element used to attach the 500-gram weight was about 2 inches. 4. Approximately every 1/2 hour, the test laminate was visually examined by quickly opening the oven door. The time at which one substrate or layer had detached from the other substrate or layer was recorded. The recorded time corresponded to the approximate time of failure of the laminate. The two, now separate, substrates were then examined to determine the nature of the failure. If the substrates separated such that most of the adhesive remained on one of the substrates, then failure was deemed to be an adhesion failure (*i.e.*, failure likely occurred at the interface between one of the substrates and the adhesive composition). If the substrates separated such that adhesive remained on both substrates, the failure was deemed to be a cohesion failure (*i.e.*, separation likely occurred within the adhesive composition itself). If neither of these conditions arose, but instead one or both of the substrates failed (*i.e.*, that portion of the laminate bonded by the adhesive, usually a 1 inch by 2 inch area of the test sample), then the failure was deemed a material failure of one or both substrates.

Dynamic Peel and Shear Testing

To determine dynamic peel strength, a laminate was tested for the maximum amount of tensile force that was needed to pull apart the layers of the laminate. Values for peel strength were obtained using a specified width of laminate (for the present application, 2 inches); clamp jaw width (for the present application, a width greater than 2 inches); and a constant rate of extension (for the present application, a rate of extension of 300 millimeters per minute). For samples having a film side, the film side of the specimen is covered with

masking tape, or some other suitable material, in order to prevent the film from ripping apart during the test. The masking tape is on only one side of the laminate and so does not contribute to the peel strength of the sample. This test uses two clamps, each clamp having two jaws with each jaw having a facing in contact with the sample, to hold the material in the same plane, usually vertically. The sample size is 2 inches (10.2 cm) wide by 4 inches (20.4 cm). The jaw facing size is 0.5 inch (1.25 cm) high by at least 2 inches (10.2 cm) wide, and the constant rate of extension is 300 mm/min. For a dynamic peel test, one clamp is attached to the top 750 of one substrate of a test panel (see Figure 7B). The other clamp is attached to the top 752 of the other substrate of a test panel. During testing, the clamps move apart at the specified rate of extension to pull apart the laminate. The sample specimen is pulled apart at 180 degrees angle of separation between the two layers, and the peel strength reported is the maximum tensile strength, in grams, recorded during the test. Each of the peel strengths reported below is an average of five to nine tests. A suitable device for determining the peel strength testing is a SINTECH 2 tester, available from the Sintech Corporation, a business having offices at 1001 Sheldon Dr., Cary, N.C. 27513; or an INSTRON Model TM, available from the Instron Corporation, a business having offices at 2500 Washington St., Canton, Mass. 02021; or the Thwing-Albert Model INTELLECTII available from the Thwing-Albert Instrument Co., a business having offices at 10960 Dutton Rd., Philadelphia, Pa. 19154.

For a dynamic shear test, the procedure is as described above except that one clamp is attached to the top 750 of one substrate of the laminate, and the other clamp is attached to the bottom 754 of the other substrate of the laminate. The shear strength reported is the maximum tensile strength, in grams, recorded during the test. Each of the shear strengths reported is an average of five to nine tests.

Accretion Value or Relative Accretion Value

The relative accretion or build-up of an adhesive, alone or in combination with other materials, e.g., fibers, was measured by running a laminate comprising adhesive through a rotary ultrasonic bonder at 300 feet per minute for ten minutes (or other specified time). The rotary bonder included a horn and a dot-pattern anvil design. The ultrasonic generator was a 3005 Autotrac, 20 KHz, 3000 watt generator from Dukane Corporation, a

business having offices in Saint Charles, Illinois. A variable-power supply was used to vary power available to the generator. The power level used was 100%, which corresponded to an ultrasonic wave amplitude of 2.8 to 3.5 mil (1 mil is equivalent to 1/1000 inch). The horn diameter was approximately 6.75 inches, with the pressure exerted by the horn on the anvil typically about 40 pounds per square inch or more to ensure good contact between the substrate, web, or laminate being processed; the horn; and the anvil.

The anvil had a dot pattern, with each pin having a 45 mil diameter and a height of 31 mil. The spacing between each pin was about 79 mil. The anvil pins were made from D2 tool steel, which was heat treated and through hardened to Rockell C 60-63. The width of the pattern was 300 mil. The diameter of the anvil was about 5.7 inches.

Additional detail on related designs and specifications pertaining to ultrasonic equipment is found in U.S. Patent Numbers 5,110,403 and 5,096,532, both of which are incorporated by reference in a manner consistent with the present application.

The build-up, which consisted of adhesive and other material, e.g., nonwoven fibers, was scraped from the horn and the anvil and weighed, giving the accretion value for the evaluated adhesive.

Laminates for this evaluation were prepared by meltblowing adhesive to get a 10 gram per square meter coverage on an approximately 0.4-ounce-per-square-yard polypropylene spunbond nonwoven facing. As shown above, adhesive was applied to one facing. This facing with the applied adhesive was then nipped together with the other facing (or substrate, in this case another 0.4 osy polypropylene spunbond substrate) to form a laminate. Typical lamination speeds were 300 feet per minute.

Conventional hot-melt adhesives that were used to prepare laminates prior to accretion-value tests included: an adhesive available under the designator H2800 from Bostik-Findley, a business having offices in Milwaukee, Wisconsin; an adhesive available under the designator H2525A from Bostik-Findley; and an adhesive available under the designator N.S.10242-94A from National Starch Co., a business having offices in Bridgewater, New Jersey.

A laminate made using a conventional hot-melt adhesive, or an adhesive of the present invention, was run through ultrasonic-bonding equipment under the conditions

described above. The accretion or buildup was scraped off the various ultrasonic-bonding surfaces after a selected time and weighed. Relative-accretion values may be calculated by dividing the accretion value of the laminate comprising an adhesive of the present invention by the accretion value of a selected conventional hot-melt adhesive (e.g., a conventional hot-melt adhesive for which an adhesive of the present invention is to be substituted).

Thermal Stability: Thermogravimetric Analysis and Differential Scanning Calorimetry

The thermal stability of versions of adhesive compositions of the present invention was determined using thermogravimetric analysis and differential scanning calorimetry. For the thermogravimetric analysis, a sample of adhesive was placed in a sample holder in the heating element of a Model 951 Thermogravimetric Analyzer made by TA Instruments, a business having offices in New Castle, Delaware. The sample was heated from room temperature, which was approximately 21°C, to a temperature of 450°C at a heating rate of 10°C per minute. The sample was heated under a dynamic atmosphere of air with a flow of approximately 80 milliliters per minute. The crucible was continuously weighed during heating so that any decrease in weight could be detected. The resulting weight-change curves for the tested adhesives, i.e. plots of sample weight versus temperature, showed that isotactic polypropylene, atactic polypropylene, and blends of atactic and isotactic polypropylene (with the blends typically ranging from about 10 weight percent to about 30 weight percent isotactic polypropylene) generally had a decomposition temperature of about 235°C in air.

For the analysis using differential-scanning calorimetry, a 10 milligram sample of isotactic polypropylene (see Example 1 below) was placed in the sample chamber of the heating/cooling block of a Model 2920 differential scanning calorimetry analyzer made by TA Instruments. The sample was heated from -100°C to 250°C, then cooled to -100°C, then reheated again to 250°C, at a heating and cooling rate of 10°C per minute. A Liquid Nitrogen Cooling Accessory, also made by TA Instruments, was attached to the Model 2920 differential scanning calorimeter. The results indicated that there was a significant peak showing energy absorption over the temperature range from about 150°C to about 170°C, with a peak at about 161°C (i.e., indicative of melting).

A 10-milligram sample of amorphous polypropylene (*see* Example 1 below) was evaluated using the same differential-scanning calorimetry procedure. The analysis indicated that the amorphous polypropylene had a glass-transition temperature of about -10 degrees Celsius.

Viscosity

Atactic and isotactic polypropylene blends of varying compositions were formulated into 10.0 g samples. These samples were heated to or above 400°F in a Brookfield Digital Rheometer Model DV-III using a Brookfield Temperature Controller (available from Brookfield Engineering Laboratories, a business having offices in Stoughton, MA). Spindle #27 was used for all trials and the instrument was appropriately zeroed and calibrated before each test. After the sample had been stabilized and sufficiently mixed at 400 degrees Fahrenheit (or above), readings of the spindle rpm, torque, and viscosity were recorded. The temperature was then lowered, typically in 10°F increments, and the sample allowed to stabilize for 10-15 minutes before subsequent readings of spindle rpm, torque, and viscosity were taken. For various blends of isotactic polypropylene and atactic polypropylene (*see* Example 1 below for characteristics), Brookfield viscosities at 360 degrees Fahrenheit were: for 10 weight percent isotactic polypropylene/90 weight percent atactic polypropylene, the viscosity was 3200 centipoise; for 20 weight percent isotactic polypropylene/80 weight percent atactic polypropylene, the viscosity was 4700 centipoise; for 30 weight percent isotactic polypropylene/70 weight percent atactic polypropylene, the viscosity was 6300 centipoise; and for 40 weight percent isotactic polypropylene/60 weight percent atactic polypropylene, the viscosity was 7000 centipoise.

For various blends of isotactic polypropylene and atactic polypropylene (*see* Example 1 below for characteristics), Brookfield viscosities at 380 degrees Fahrenheit were: for 10 weight percent isotactic polypropylene/90 weight percent atactic polypropylene, the viscosity was 2500 centipoise; for 20 weight percent isotactic polypropylene/80 weight percent atactic polypropylene, the viscosity was 3600 centipoise; for 30 weight percent isotactic polypropylene/70 weight percent atactic polypropylene, the viscosity was 4900 centipoise; and for 40 weight percent isotactic polypropylene/60 weight percent atactic polypropylene, the viscosity was 5300 centipoise.

Molecular Weight (Number Average and Weight Average)

Atactic polypropylene, isotactic polypropylene, and blends of atactic and isotactic polypropylene were sent to American Polymer Standard Corp., a business having offices in Philadelphia, PA, for molecular-weight determinations. The number-average and/or weight-average molecular weights were determined by American Polymer using gel-permeation chromatography on a Waters Model No. 150 gel-permeation chromatograph. The determinations were made using: four, linear, Shodex GPC gel columns; poly(styrene-divinyl benzene) copolymers as standards; trichlorobenzene as the solvent, introduced to the chromatograph at a volumetric flow rate of 1.0 milliliter per minute; an operating temperature of 135 degrees Celsius; a sample-injection volume of 100 microliters; an M-150C-(64/25) detector; and a GPC PRO 3.13 IBM AT data module.

EXAMPLESExample 1

Amorphous (*i.e.*, atactic) polypropylene (received as slightly yellowish, transparent, elastomeric, small blocks) was obtained from Sigma-Aldrich, a business having offices in Milwaukee, Wisconsin. The amorphous polypropylene had a weight-average molecular weight of about 28,000 and a number-average molecular weight of 8400 (determined by American Polymer as discussed above). Furthermore, the procured amorphous polypropylene was determined to have a Brookfield viscosity of 23 poise at a temperature of 190 degrees Celsius (determine as described in the Tests/Procedures section above); and a glass-transition temperature of -10 degrees Celsius (determined, as described above, using differential-scanning calorimetry at a heating rate of 10 degrees Celsius per minute).

Isotactic polypropylene was obtained from Sigma-Aldrich in the form of white, spherical particles. The isotactic polypropylene was determined to have a number-average molecular weight of about 20,000 and a weight-average molecular weight of about 110,000. The procured isotactic polypropylene had a melting index of 1000 grams per ten minutes (at a temperature of 230 degrees Celsius and when subjected to a force of 2.16 kg; *see* ASTM D 1238, which was used for this determination, for additional detail on measuring the melting index).

Four and 1/2 kilograms of amorphous polypropylene were added to an adhesive-supply unit; *i.e.*, a hot-melt processing system comprising a hot-melt tank. The adhesive-supply unit was a Dynamelt S Series Model Number DM310, available from ITW Dynatec, a business having offices in Hendersonville, Tennessee. The adhesive-supply unit was set at a temperature of about 370 degrees Fahrenheit. After about one hour the amorphous polypropylene was in substantially liquid form. One-half kilogram of isotactic polypropylene was then added to the already-liquefied amorphous polypropylene. To ensure adequate mixing and uniformity of the resulting blend, a circulatory pump associated with the adhesive-supply unit was activated. The circulatory pump, a gear pump, was operated so that approximately 0.5 to 0.8 pounds per minute of the precursor materials to the uniform blend were circulated. The pump was run for about 1 hour, the time at which the isotactic polypropylene solids were observed to have melted and become uniformly blended with the liquefied amorphous polypropylene.

The Brookfield viscosity of the blend was determined to be about 3000 centipoise at a temperature of 370 degrees Celsius.

Example 2

A number of adhesives were used to bond two substrates together to make laminated structures using the procedures described above. The integrity of the bond between the two substrates was then tested using the static-peel test described above. In one evaluation, two necked-bonded laminate (NBL) substrates were bonded together. For additional detail on how NBLs and other neck-bonded materials are formed, see U.S. Patent Number 5,336,545 to Morman, entitled "Composite Elastic Necked-Bonded Material," which is hereby incorporated by reference in its entirety in a manner consistent with the present document.

Laminates comprising two adhesively-bonded NBL substrates were made and tested as described above. Each laminate comprised a first NBL substrate, as described above, which was adhesively bonded to a second NBL substrate. For this particular example, each of the adhesives was slot-coated on to one of the substrate layers when making the laminate. Each adhesive was applied at an add-on level of about 20 grams per square meter. The same NBL substrate was used to make all laminates in this Example. The following

comparative adhesive compositions were tested: (1) three hot-melt adhesive compositions identified by the alpha-numeric designators N.S. 8819-6C, N.S. 11414-19A, and N.S. 11414-19D, which are available from National Starch, a business having offices in Bridgewater, New Jersey; (2) three hot-melt adhesive compositions identified by the alpha-numeric designators H8163, H2800, and H2727, which are all available from Bostik-Findley, a business having offices in Milwaukee, Wisconsin; and (3) an amorphous poly-alpha-olefin hot-melt adhesive, comprising a poly(ethylene-co-propylene) copolymer, which is available under the designator RT2730 from Huntsman Polymer Corp., Houston, Texas.

For this example, we made an embodiment of our invention by blending amorphous (*i.e.*, atactic) polypropylene and isotactic polypropylene as described in Example 1 above. The static peel strength was determined as described above (*i.e.*, a 500 gram mass was attached to the upper edge of one of the substrates of a test panel, with the test laminate suspended in an oven at a temperature of 100 degrees Fahrenheit). Results of this comparison are provided in Table 1 below:

Table 1: Comparison of Adhesive Performance

Adhesive Type	Time of Detachment Between Two Substrates (<i>i.e.</i> , Failure)	Nature of Failure
N.S. 8819-6C	2 hours or less	Cohesion
N.S. 11414-19A	1 hour or less	Cohesion
N.S. 11414-19D	1 hour or less	Cohesion
H8163	4 hours or less	Cohesion
H2800	4 hours or less	Cohesion
H2727	4 hours or less	Cohesion
RT2730	1 hour or less	Cohesion
Atactic/isotactic Polypropylene Blend (10% by weight isotactic polypropylene)	No bond failure after 30 hours	Necked-bonded laminate failed (<i>i.e.</i> , the NBL substrate itself delaminated)

Example 3

Laminates were made with approximately 0.4 osy polypropylene, spunbonded substrates and adhesive applied in a meltblown pattern at an application level of about 10 grams per square meter. The laminate was then run through ultrasonic-bonding equipment using the procedures described above to determine the accretion value for each of the tested adhesives.

For a laminate made using the blend of atactic polypropylene and isotactic polypropylene described in Example 1, no buildup or residue was observed on the surfaces of the anvil and horn after the ultrasonic-bonding equipment had been run for approximately 8 and ½ minutes.

For a laminate made using the H2800 hot-melt adhesive, about 0.4 to about 0.5 grams of buildup or residue was collected from the surfaces of the anvil and horn after the ultrasonic-bonding equipment had been run for approximately 8 and 1/2 minutes.

For a laminate made using the H2525A hot-melt adhesive, about 1.8 grams of buildup or residue was collected from the surfaces of the anvil and horn after the ultrasonic-bonding equipment had been run for approximately 5 minutes.

For a laminate made using the N.S.10242-94A hot-melt adhesive, about 1.2 grams of buildup or residue were collected from the surfaces of the anvil and horn after the ultrasonic-bonding equipment had been run for approximately 5 minutes. Example 4

An embodiment of an adhesive composition of the present invention was used to bond two substrates together. For this Example, one of the substrates was a necked-bonded laminate substrate as discussed above. The second substrate was an outer cover as described above.

A laminate was made by adhesively bonding the NBL substrate to the outer cover material as described above. The adhesive was applied, and the NBL substrate and outer cover were nipped together in a way such that the polypropylene, spunbonded layer of the outer cover contacted the adhesive. A version of an adhesive composition of the present invention was made as discussed in Example 1 above. Specifically a blend of atactic and isotactic polypropylene was prepared and used in making the laminate described in this Example (with the isotactic polypropylene present as 10% by weight of the total weight of

the blend). The adhesive was slot coated onto one of the substrates at 20 grams per square meter before the NBL substrate and outer cover were nipped together with the adhesive located between the two substrates. The static peel strength was determined as described above (*i.e.*, a 500 gram mass was attached to the upper edge of one of the substrates, with the test panel suspended in an oven at a temperature of 100 degrees Fahrenheit). For a laminate comprising an adhesive of the present invention, the time at which one substrate detached from the other exceeded 24 hours. Again, there was a material failure, the NBL substrate itself delaminated, not a bond failure.

A second comparative laminate was made as described above (*i.e.* an NBL substrate was adhesively bonded to an outer-cover substrate, with each of the substrates being the same as those used to prepare a laminate comprising an adhesive of the present invention as described in the preceding paragraph) except an embodiment of an adhesive composition of the present invention was replaced with RT2730 available from Huntsman Polymer Corp., as described above. The adhesive was slot coated onto one of the substrates at 20 grams per square meter before the NBL substrate and outer cover were nipped together with the adhesive located between the two substrates. The static peel strength was determined as described above (*i.e.*, a 500 gram mass was attached to the upper edge of one of the substrates, with the test panel suspended in an oven at a temperature of 100 degrees Fahrenheit). The time at which the substrates detached from one another was less than 1 hour. The nature of the detachment was a cohesive failure of the adhesive bond.

Example 5

A laminate was made using a necked-bonded-laminate substrate (*i.e.*, an NBL substrate) and an outercover material (as described above). The adhesive was applied, and the NBL substrate and outer cover were nipped together in a way such that the polypropylene, spunbonded layer of the outer cover contacted the adhesive. A version of an adhesive composition of the present invention was made as discussed in Example 1 above. Specifically a blend of atactic and isotactic polypropylene was prepared and used in making the laminate described in this Example (with the isotactic polypropylene present as 15% by weight of the total weight of the blend). The adhesive was meltblown onto one of the substrates at either 10 grams per square meter or 20 grams per square meter, as described

below, before the NBL substrate and outer cover were nipped together with the adhesive located between the two substrates. The static peel strength was determined as described above (*i.e.*, a 500 gram mass was attached to the upper edge of one of the substrates, with the test panel suspended in an oven at a temperature of 100 degrees Fahrenheit). For a laminate comprising an adhesive of the present invention, the time at which one substrate detached from the other exceeded 24 hours. Again, there was a material failure, the NBL substrate itself delaminated, not a bond failure. The dynamic-peel-test value for this same laminate, determined using the procedures discussed above, was about 870 grams per inch. The mode of failure for this dynamic test was a material failure (*i.e.*, delamination of the NBL substrate).

A second laminate was made in the same way except that the adhesive of the present invention was applied at an add-on level of about 20 grams per square meter. The dynamic-peel-test value for this second laminate, prepared at a higher add-on level, was determined to be about 1160 grams per inch. The mode of failure for this dynamic test was a material failure (*i.e.*, delamination of the NBL substrate).

The dynamic-shear-test value was also determined for each laminate. At an add-on level of 10 grams per square meter, the dynamic-shear-test value was about 2170 grams per square inch. At an add-on level of 20 grams per square meter, the dynamic-shear-test value was about 2190 grams per square inch. The mode of failure for both of these dynamic shear tests was a material failure (*i.e.*, delamination of the NBL substrate).

Example 6

A laminate was made using a necked-bonded-laminate substrate (*i.e.*, an NBL substrate) and a stretch-bonded laminate ("SBL") as a substrate. An SBL is generally a laminate made up of an elongated elastic web or elongated elastomeric strands bonded between two spunbond layers, for example. For additional detail on how SBLs are formed, see European Patent Application No. EP 0 217 032 published on April 8, 1987 in the names of Taylor et al., which is hereby incorporated by reference in its entirety in a manner consistent with the present document.

The adhesive was applied, and the NBL substrate and SBL substrate were nipped together with the adhesive now located between the substrates. A version of an

adhesive composition of the present invention was made as discussed in Example 1 above, except that the composition was made so that isotactic polypropylene was present as 15% by weight of the total weight, i.e., the adhesive composition comprised 15 weight percent isotactic polypropylene. The adhesive was meltblown onto one of the substrates at either 20 grams per square meter or 40 grams per square meter, as described below, before the NBL substrate and SBL substrate were nipped together with the adhesive located between the two substrates. The dynamic-peel-test value for this NBL/SBL laminate comprising a 15-weight-percent isotactic polypropylene/85-weight-percent atactic polypropylene blend applied at 20 grams per square meter was determined to be 1220 grams per inch. The mode of failure for this dynamic test was a material failure (i.e., delamination of the NBL substrate and SBL substrate). The dynamic-shear-test value for this NBL/SBL laminate comprising a 15-weight-percent isotactic polypropylene/85-weight-percent atactic polypropylene blend applied at 20 grams per square meter was determined to be 2540 grams per square inch. The mode of failure for this dynamic shear test was a material failure (i.e., delamination of the NBL substrate).

The dynamic-peel-test value for this NBL/SBL laminate comprising a 15-weight-percent isotactic polypropylene/85-weight-percent atactic polypropylene blend applied at 40 grams per square meter was determined to be 1580 grams per inch. The mode of failure for this dynamic test was a material failure (i.e., delamination of the NBL substrate and SBL substrate). The dynamic-shear-test value for this NBL/SBL laminate comprising a 15-weight-percent isotactic polypropylene/85-weight-percent atactic polypropylene blend applied at 40 grams per square meter was determined to be 2280 grams per square inch. The mode of failure for this dynamic shear test was a material failure (i.e., delamination of the NBL substrate).

Another laminate was made in the same way except that the adhesive of the present invention was replaced with a conventional hot-melt adhesive available under the designator H2800 from Bostik-Findley. This adhesive was meltblown at an add-on level of 40 grams per square meter. The dynamic-peel-test value for this NBL/SBL laminate comprising the H2800 adhesive, applied at 40 grams per square meter, was determined to be 1470 grams per inch. The mode of failure for this dynamic test was a material failure (i.e.,

delamination of the NBL substrate and SBL substrate). The dynamic-shear-test value for this NBL/SBL laminate comprising the H2800 adhesive, applied at 40 grams per square meter, was determined to be 2800 grams per square inch. The mode of failure for this dynamic shear test was a material failure (*i.e.*, delamination of the NBL substrate).

5 It will be appreciated that details of the foregoing embodiments, given for purposes of illustration, are not to be construed as limiting the scope of this invention. Although only a few exemplary embodiments of this invention have been described in detail above, those skilled in the art will readily appreciate that many modifications are possible in the exemplary embodiments without materially departing from the novel teachings and advantages of this invention. Accordingly, all such modifications are intended to be included
10 within the scope of this invention, which is defined in the following claims and all equivalents thereto. Further, it is recognized that many embodiments may be conceived that do not achieve all of the advantages of some embodiments, particularly of the preferred embodiments, yet the absence of a particular advantage shall not be construed to necessarily
15 mean that such an embodiment is outside the scope of the present invention.

WHAT IS CLAIMED IS:

1. An adhesive composition, comprising:
an atactic polymer having a degree of crystallinity of less than about 20% and
a number-average molecular weight between about 1,000 and about 300,000; and
an isotactic polymer having a degree of crystallinity of at least about 40% and
a number-average molecular weight between about 3,000 and about 200,000.
2. The adhesive composition of Claim 1, wherein the degree of crystallinity of the atactic polymer is less than about 15%.
3. The adhesive composition of Claim 1, wherein the degree of crystallinity of the isotactic polymer is at least about 60%.
4. The adhesive composition of Claim 1, wherein the degree of crystallinity of the isotactic polymer is at least about 80%.
5. The adhesive composition of Claim 1, wherein the isotactic polymer is at least 60% isotactic.
6. The adhesive composition of Claim 1, wherein the isotactic polymer is at least 70% isotactic.
7. The adhesive composition of Claim 1, wherein the isotactic polymer is at least 80% isotactic.
8. The adhesive composition of Claim 1, wherein the number-average molecular weight of the atactic polymer is between about 3,000 and about 100,000.

9. The adhesive composition of Claim 1, wherein the number-average molecular weight of the isotactic polymer is between about 10,000 and about 100,000.

10. The adhesive composition of Claim 1, wherein the adhesive composition is hot-melt processable at less than about 450 degrees Fahrenheit.

11. The adhesive composition of Claim 1, wherein the adhesive composition is hot-melt processable at less than about 400 degrees Fahrenheit.

12. The adhesive composition of Claim 1, wherein the adhesive composition is hot-melt processable at less than about 375 degrees Fahrenheit.

13. The adhesive composition of Claim 1, wherein the adhesive composition is hot-melt processable at less than about 350 degrees Fahrenheit.

14. The adhesive composition of Claim 1, wherein the adhesive composition has a melt index between about 100 and about 2000 grams per 10 minutes.

15. The adhesive composition of Claim 1, wherein the adhesive composition has a melt index between about 200 and about 1800 grams per 10 minutes.

16. The adhesive composition of Claim 1, wherein the adhesive composition has a melt index between about 500 and about 1500 grams per 10 minutes.

17. The adhesive composition of Claim 1, comprising between about 50 and about 90 weight percent of the atactic polymer, and between about 5 and about 50 weight percent of the isotactic polymer.

18. The adhesive composition of Claim 1, wherein the atactic polymer comprises atactic polypropylene.

19. The adhesive composition of Claim 1, wherein the atactic polymer is selected from the group consisting of: low density polyethylene, atactic polystyrene, atactic polybutene, amorphous polyolefin copolymer, and combinations thereof.

20. The adhesive composition of Claim 19, wherein the low density polyethylene has a density in a range of 0.910 to 0.935 grams per cubic centimeter.

21. The adhesive composition of Claim 1, wherein the isotactic polymer comprises isotactic polypropylene.

22. The adhesive composition of Claim 1, wherein the isotactic polymer is selected from the group consisting of: high density polyethylene, isotactic polystyrene, isotactic polybutene, and combinations thereof.

23. The adhesive composition of Claim 22, wherein the high density polyethylene has a density in a range of 0.935 to 0.980 grams per cubic centimeter.

24. A laminated structure comprising at least a portion of a first layer attached to at least a portion of a second layer using the adhesive composition of Claim 1.

25. The laminated structure of Claim 24, wherein the laminated structure has a static-peel-failure time of at least about 1 hour.

26. The laminated structure of Claim 24, wherein the laminated structure has a static-peel-failure time of at least about 8 hours.

27. The laminated structure of Claim 24, wherein the laminated structure has a static-peel-failure time of at least about 24 hours.

28. The laminated structure of Claim 24, wherein the laminated structure has a relative accretion value of less than 1.

29. The laminated structure of Claim 24, wherein the laminated structure has a relative accretion value of less than 0.5.

30. The laminated structure of Claim 24, wherein the laminated structure has a relative accretion value of less than 0.2.

31. The laminated structure of Claim 24, wherein the first and second layers are each part of a single substrate.

32. The laminated structure of Claim 24, wherein each of the first and second layers is selected from the group consisting of: nonwoven material, woven material, film, and an elasticized component.

33. The laminated structure of Claim 24, wherein at least one of the first and second layers comprises at least one of the group consisting of cellulosic material, thermoplastic material, and combinations thereof.

34. An absorbent article comprising the laminated structure of Claim 24.

35. The adhesive composition of Claim 1, further comprising up to 50% by weight of a combination of additives selected from the group consisting of: a tackifier, an antioxidizing agent, color pigment, filler, and a polymer compatibilizer, wherein the adhesive composition has an open time of up to 2 minutes.

36. The adhesive composition of Claim 35, wherein the adhesive composition has an open time of up to 30 seconds.

37. The adhesive composition of Claim 35, wherein the adhesive composition has an open time of up to 10 seconds.

38. The adhesive composition of Claim 35, wherein the adhesive composition has an open time of up to 1 second.

39. An absorbent article comprising the adhesive composition of Claim 1.

40. An adhesive composition, comprising:
at least 50% and less than 80% by weight of an atactic polymer having a degree of crystallinity of less than about 20% and a number-average molecular weight of at least 3,000 and less than 100,000; and
between about 5% and about 40% by weight of an isotactic polymer having a degree of crystallinity of at least about 40%.

41. The adhesive composition of Claim 40, wherein the degree of crystallinity of the atactic polymer is less than about 15%.

42. The adhesive composition of Claim 40, wherein the degree of crystallinity of the isotactic polymer is at least about 60%.

43. The adhesive composition of Claim 40, wherein the degree of crystallinity of the isotactic polymer is at least about 80%.

44. The adhesive composition of Claim 40, wherein the isotactic polymer is at least 60% isotactic.

45. The adhesive composition of Claim 40, wherein the isotactic polymer is at least 70% isotactic.

46. The adhesive composition of Claim 40, wherein the isotactic polymer is at least 80% isotactic.

47. The adhesive composition of Claim 40, wherein the number-average molecular weight of the isotactic polymer is between about 3,000 and about 200,000.

48. The adhesive composition of Claim 40, wherein the number-average molecular weight of the isotactic polymer is between about 10,000 and about 100,000.

49. The adhesive composition of Claim 40, wherein the adhesive composition has a melt index between about 100 and about 2000 grams per 10 minutes.

50. The adhesive composition of Claim 40, wherein the adhesive composition has a melt index between about 200 and about 1800 grams per 10 minutes.

51. The adhesive composition of Claim 40, wherein the adhesive composition has a melt index between about 500 and about 1500 grams per 10 minutes.

52. The adhesive composition of Claim 40, wherein the atactic polymer comprises atactic polypropylene.

53. The adhesive composition of Claim 40, wherein the atactic polymer is selected from the group consisting of: low density polyethylene, atactic polystyrene, atactic polybutene, amorphous polyolefin copolymer, and combinations thereof.

54. The adhesive composition of Claim 53, wherein the low density polyethylene has a density in a range of 0.910 to 0.935 grams per cubic centimeter.

55. The adhesive composition of Claim 40, wherein the isotactic polymer comprises isotactic polypropylene.

56. The adhesive composition of Claim 40, wherein the isotactic polymer is selected from the group consisting of: high density polyethylene, isotactic polystyrene, isotactic polybutene, and combinations thereof.

57. The adhesive composition of Claim 56, wherein the high density polyethylene has a density in a range of 0.935 to 0.980 grams per cubic centimeter.

58. A method of making an adhesive composition, comprising the steps of:

providing an atactic polymer having a degree of crystallinity of less than about 20% and a number-average molecular weight between about 1000 and about 300,000;

providing an isotactic polymer having a degree of crystallinity of at least about 40% and a number-average molecular weight between about 3000 and about 200,000;

heating the atactic polymer and the isotactic polymer until the atactic polymer and the isotactic polymer are liquified;

blending the heated atactic polymer and the heated isotactic polymer to form an adhesive composition that is melt-processable at a temperature of less than about 450 degrees Fahrenheit.

59. The method of Claim 58, wherein the atactic polymer comprises atactic polypropylene.

60. The method of Claim 58, wherein the atactic polymer is selected from the group consisting of: low density polyethylene, atactic polystyrene, atactic polybutene, amorphous polyolefin copolymer, and combinations thereof.

61. The method of Claim 60, wherein the low density polyethylene has a density in a range of 0.910 to 0.935 grams per cubic centimeter.

62. The method of Claim 58, wherein the isotactic polymer comprises isotactic polypropylene.

63. The method of Claim 58, wherein the isotactic polymer is selected from the group consisting of: high density polyethylene, isotactic polystyrene, isotactic polybutene, and combinations thereof.

64. The method of Claim 63, wherein the high density polyethylene has a density in a range of 0.935 to 0.980 grams per cubic centimeter.

65. The method of Claim 58, wherein the adhesive composition comprises between about 50 and about 90 weight percent of the atactic polymer, and between about 5 and about 50 weight percent of the isotactic polymer.

66. The method of Claim 58, further comprising the steps of:
providing a first substrate;
providing a second substrate;
applying the adhesive composition to at least one of the first substrate and the second substrate; and
joining at least a portion of the first substrate to at least a portion of the second substrate with at least a portion of the applied adhesive composition positioned between the first substrate and second substrate.

67. The method of Claim 66, wherein the first and second substrates are each part of a single substrate.

68. The method of Claim 66, wherein each of the first and second substrates is selected from the group consisting of: nonwoven material, woven material, film, and an elasticized component.

69. The method of Claim 66, wherein at least one of the first and second substrates comprises at least one of the group consisting of cellulosic material, thermoplastic material, and combinations thereof.

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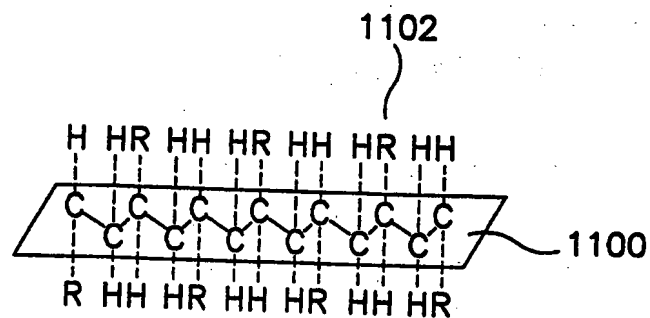


FIG. 1A

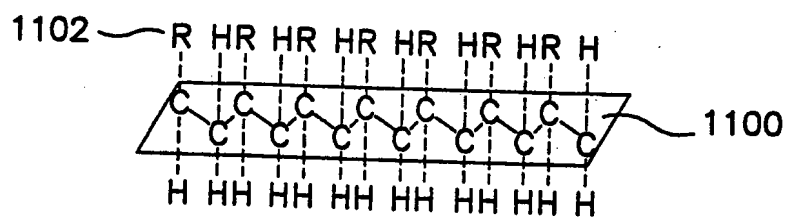


FIG. 1B

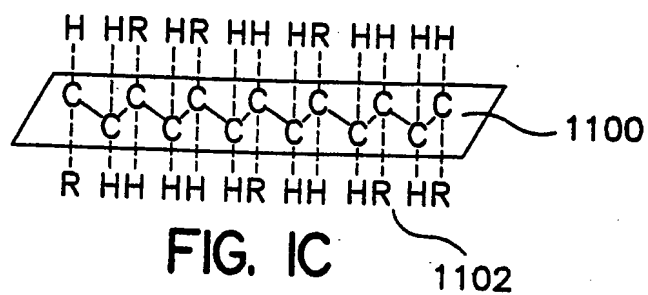


FIG. 1C

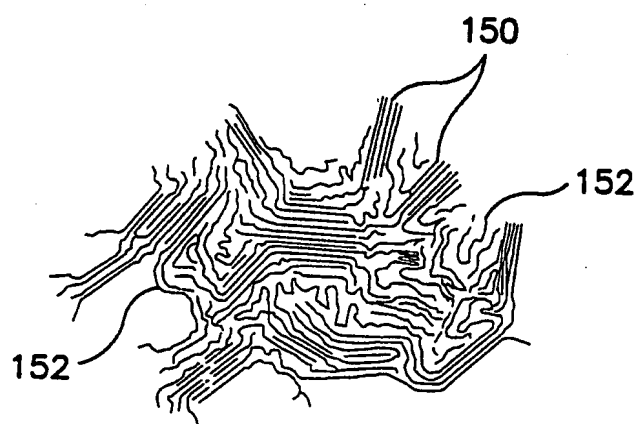


FIG. 2

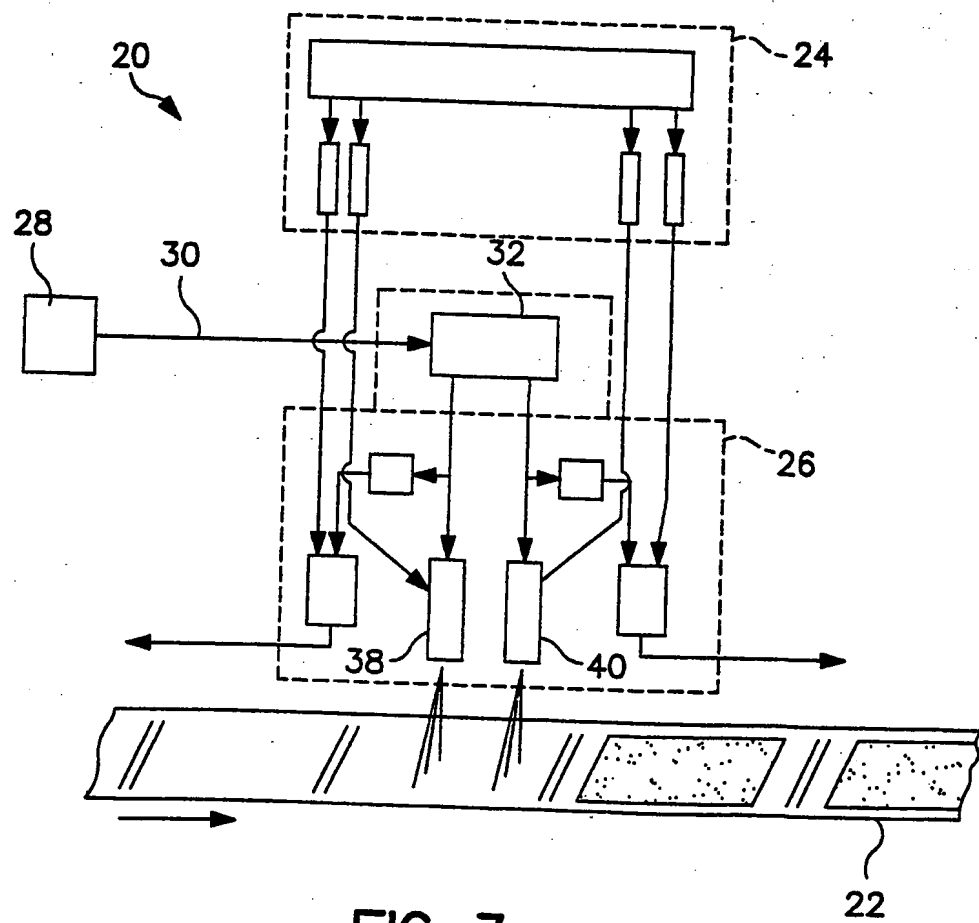


FIG. 3

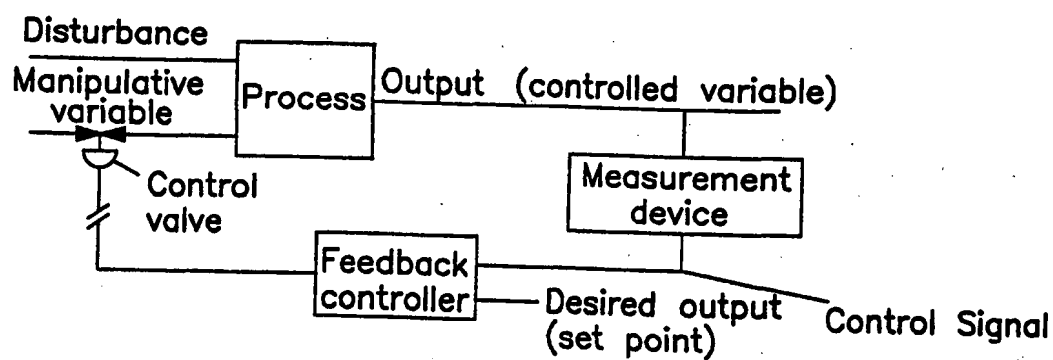


FIG. 4A

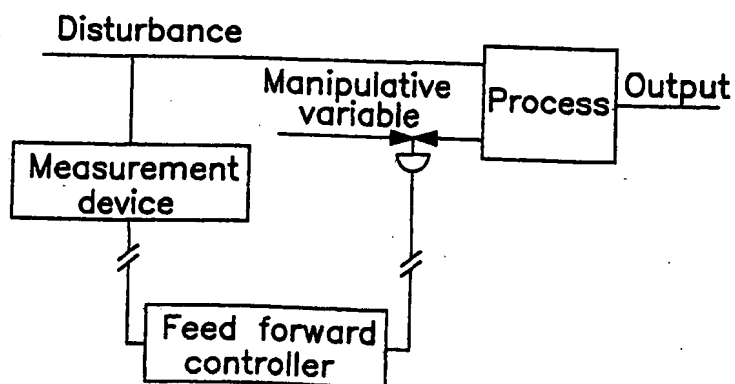


FIG. 4B

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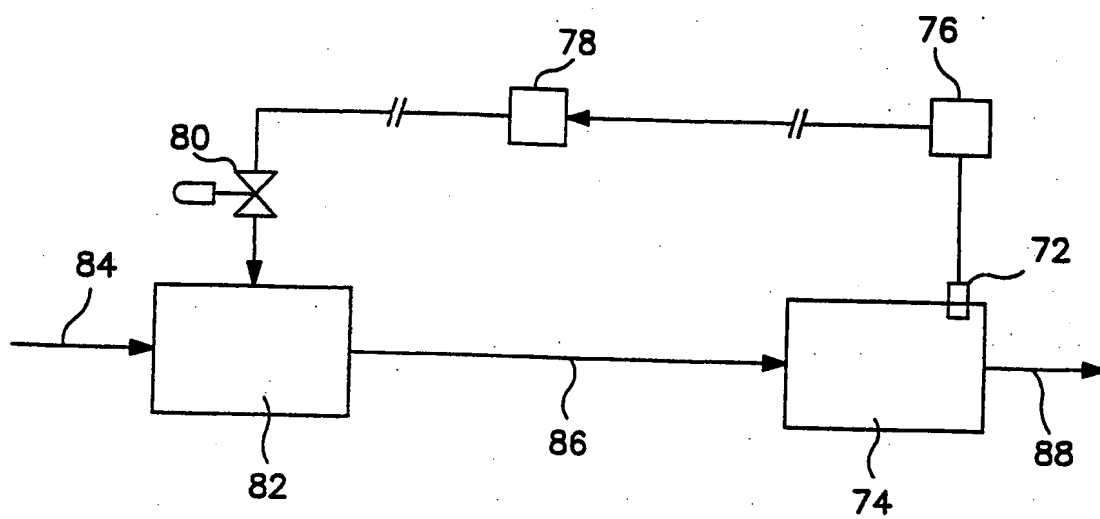


FIG. 5

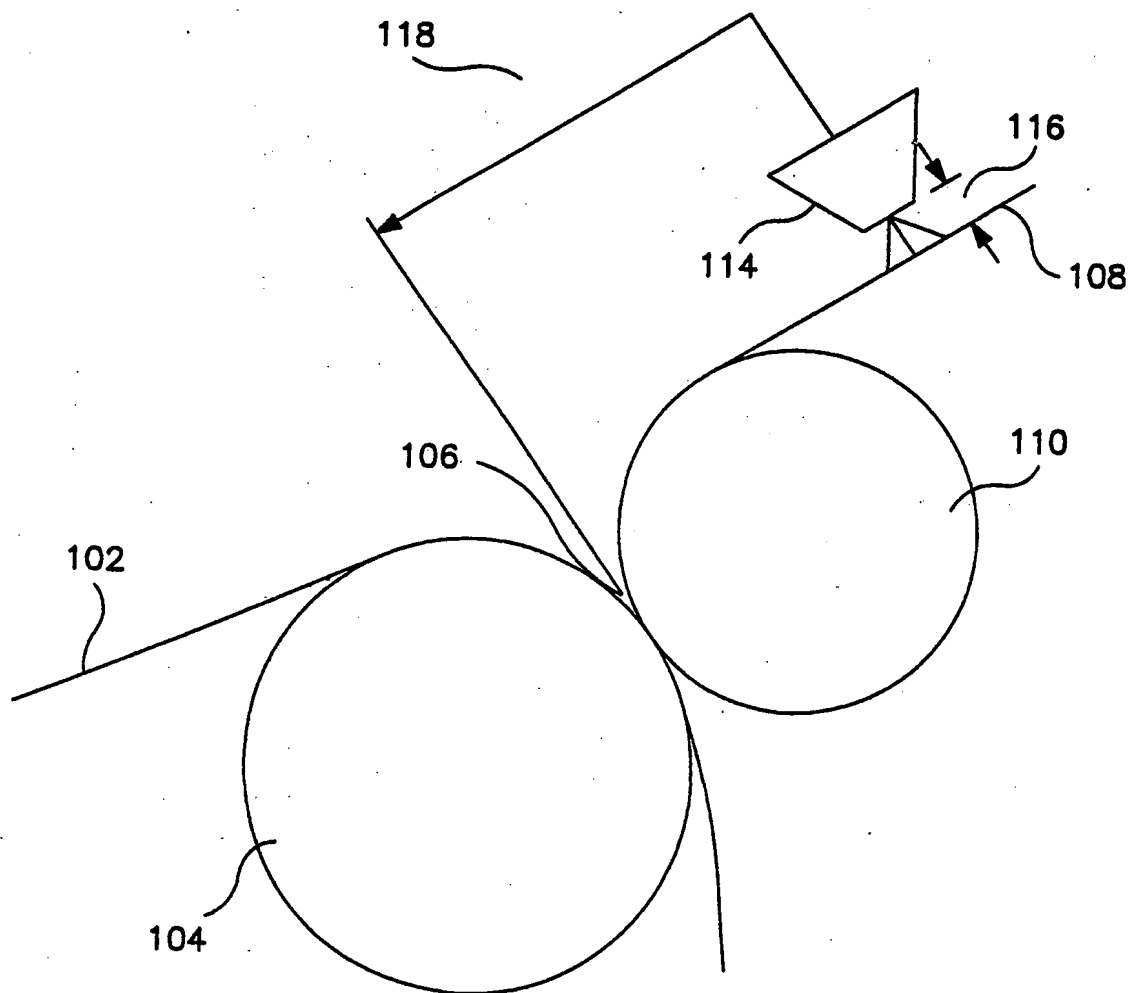


FIG. 6

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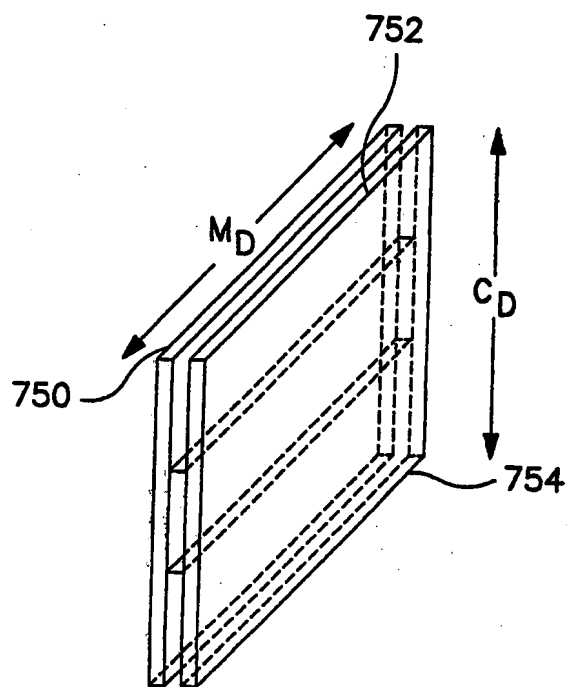


FIG. 7A

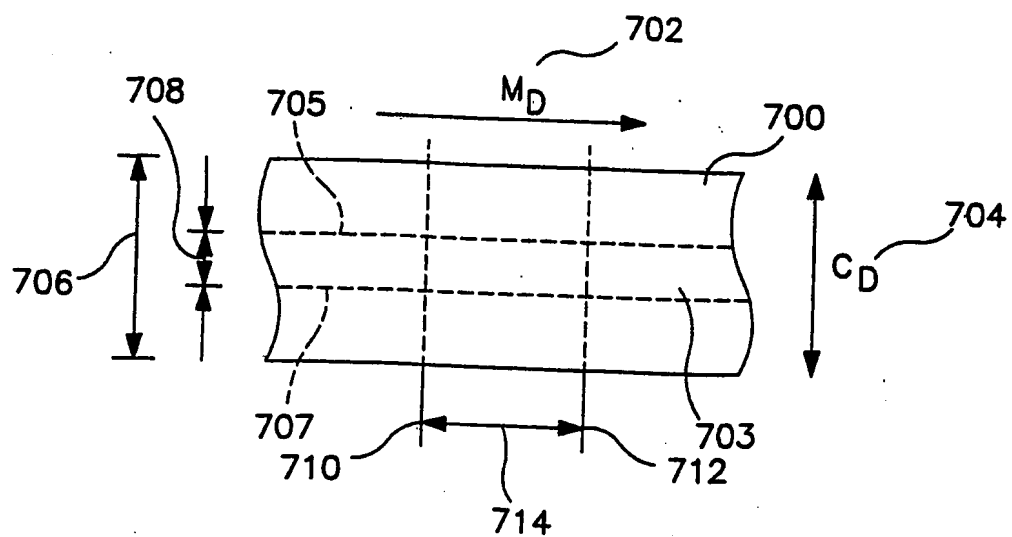


FIG. 7B

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- as to the applicant's entitlement to claim the priority of the earlier application (Rule 4.17(iii)) for all designations

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(54) Title: **HOT-MELT ADHESIVE BASED ON BLEND OF AMORPHOUS AND CRYSTALLINE POLYMERS FOR MULTI-LAYER BONDING**

(57) Abstract: The present invention is generally directed to adhesive compositions comprising selected ratios of crystalline and amorphous polymers. In some versions of the invention, polymers capable of existing in different configurations (e.g., a polymer such as polypropylene which can exist in an atactic, syndiotactic, or isotactic configuration) is used to prepare adhesives of the present invention. As an example, a selected amount of isotactic polypropylene is blended with a selected amount of atactic polypropylene to prepare an adhesive composition having one or more performance properties (e.g., bond strength) that are superior to the performance properties of a conventional hot-melt adhesive.



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INTERNATIONAL SEARCH REPORT

International Application No
CT/US 01/48241

A. CLASSIFICATION OF SUBJECT MATTER

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According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 325 562 A (PETERKIN MELVIN E) 13 June 1967 (1967-06-13)	1-69
Y	claim 2	24-34
X	US 3 370 106 A (BOYER JACKSON S ET AL) 20 February 1968 (1968-02-20) example 1	1-69
X	US 5 539 056 A (YANG HENRY W ET AL) 23 July 1996 (1996-07-23) column 28, line 4; example A	1-69
Y	WO 96 06966 A (KIMBERLY CLARK CO) 7 March 1996 (1996-03-07) examples 1,2	24-34
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 99 14039 A (KIMBERLY CLARK CO) 25 March 1999 (1999-03-25) column 10, line 13 - line 21 -----	24-34

INTERNATIONAL SEARCH REPORT

Information on patent family members

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Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 3325562	A	13-06-1967	US 3341626 A	12-09-1967
			US 3370106 A	20-02-1968
			US 3356766 A	05-12-1967
			US 3360488 A	26-12-1967
			DE 1594261 A1	01-10-1970
			DE 1594263 A1	18-02-1971
			FR 1475072 A	31-03-1967
			GB 1075299 A	12-07-1967
			NL 6603073 A	12-09-1966
			NL 6608975 A	23-01-1967
US 3370106	A	20-02-1968	US 3341626 A	12-09-1967
			US 3325562 A	13-06-1967
			US 3356766 A	05-12-1967
			US 3360488 A	26-12-1967
			DE 1594261 A1	01-10-1970
			DE 1594263 A1	18-02-1971
			FR 1475072 A	31-03-1967
			GB 1075299 A	12-07-1967
			NL 6603073 A	12-09-1966
			NL 6608975 A	23-01-1967
US 5539056	A	23-07-1996	CA 2209773 A1	08-08-1996
			DE 69607147 D1	20-04-2000
			DE 69607147 T2	28-09-2000
			EP 0807143 A1	19-11-1997
			ES 2144725 T3	16-06-2000
			JP 11500474 T	12-01-1999
			WO 9623838 A1	08-08-1996
			US 5955625 A	21-09-1999
WO 9606966	A	07-03-1996	AU 691644 B2	21-05-1998
			AU 3202495 A	22-03-1996
			BR 9508660 A	18-11-1997
			CA 2146064 A1	01-03-1996
			DE 19581747 T0	17-07-1997
			GB 2307488 A ,B	28-05-1997
			WO 9606966 A1	07-03-1996
WO 9914039	A	25-03-1999	US 6096668 A	01-08-2000
			AU 735677 B2	12-07-2001
			AU 9390398 A	05-04-1999
			BR 9812205 A	18-07-2000
			CA 2301002 A1	25-03-1999
			CN 1270552 T	18-10-2000
			EP 1015241 A1	05-07-2000
			WO 9914039 A1	25-03-1999